SUDBURY ENVIRONMENTAL STUDY

EMISSIONS OF SULPHUR OXIDES, PARTICULATES AND TRACE ELEMENTS IN THE SUDBURY BASIN.

SES 008/82

FALL 1982

Ontario
Ministry of the Environment

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Deputy Minister

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October 1982

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SES 008/82
ISBN 0-7743-7270-2
ARB-ETRD-09-82

TABLE OF CONTENTS

			Page
List of Tables			i
List of Figures			ii
Acknowledgements			iii
1.	Introduction		1
2.	Summary		5
3.	Description of Sources and Processes		9
	3.1	General	9
	3.2	The Inco 381 m Stack and the Smelter Building at Copper Cliff	9
		3.2.1 Copper Circuit	10
		3.2.2 Nickel Circuit	11
	3.3	The Inco 194 m Stack and the Iron Ore Recovery Plant Complex at Copper Cliff	12
		3.3.1 Iron Ore Recovery and Pelletizer Plants	13
		3.3.2 Nickel Refinery	14
		3.3.3 Sulphuric Acid Plants	15
	3.4	The Falconbridge 93 m Stack	15
4.	Discussion of Emission Measurement Methods, Their Frequency and Accuracy		17
5.	Sulphur Dioxide Emissions		22
	5.1	The Inco 381 m Stack	23
	5.2	Inco Low Level Emissions	28
	5.3	The Inco 194 m Stack	29
	5.4	The Falconbridge 93 m Stack	30

TABLE OF CONTENTS - Continued

		Page
6.	Emissions and Size of Total Particulates	32
6.1	The Inco 381 m Stack	33
6.2	The Inco 194 m Stack	36
6.3	The Falconbridge 93 m Stack	40
6.4	Inco's Low Level Emissions	41
6.5	Size of Emitted Particulates	42
6.5.1	Particulate Size Measurements at the Inco 381 m Stack	43
6.5.2	Particulate Size Measurements at the Falconbridge 93 m Stack	44
7.	Emissions of Metals	46
7.1	The Inco 381 m Stack	48
7.2	The Inco 194 m Stack	58
7.3	Low Level Emission from the Inco Smelter Building	60
7.4	The Falconbridge 93 m Stack	61
8.	Emissions of Sulphuric Acid	65
9.	References	67

LIST OF TABLES

	TITLE	PAGE
1.	AVERAGE YEARLY EMISSIONS OF MAJOR POLLUTANTS IN THE SUDBURY BASIN IN TONNES FOR THE PERIOD 1973-1981	6
2.	YEARLY EMISSIONS OF SULPHUR DIOXIDE FROM MAJOR INDUSTRIAL SOURCES IN THE SUDBURY BASIN IN THOUSAND TONNES	23
3.	SULPHUR DIOXIDE EMISSIONS FROM INDIVIDUAL PROCESSES TO THE INCO 194 METRE STACK IN TONNES PER/DAY IN 1977	30
4.	YEARLY EMISSIONS OF TOTAL PARTICULATES IN TONNES FROM MAJOR INDUSTRIAL SOURCES IN THE SUDBURY BASIN FROM 1973 TO 1981	33
5.	MEASURED AVERAGE PARTICULATE EMISSIONS FROM THE INCO 381 M STACK IN TONNES/HOUR	35
6.	PARTICULATE EMISSIONS FROM THE INCO 194 M STACK IN TONNES/DAY	39
7.	SUMMARY OF PARTICLE SIZE MEASUREMENTS AT THE INCO 381 M STACK	44
8.	SIZE OF PARTICULATES EMITTED FROM THE FALCONBRIDGE 93 M STACK	45
9.	AVERAGE EMISSIONS OF METALS FROM MAJOR SUDBURY SOURCES IN KILOGRAMS/HOUR	47
10.	AVERAGE EMISSIONS OF FIVE MAJOR METALS FROM THE INCO 381 M STACK IN KILOGRAM/HOUR	49
11.	AVERAGE MEASURED EMISSIONS OF METALS AND OTHER PARTICULATE POLLUTANTS FROM THE INCO 381 M STACK IN KILOGRAMS/HOUR	57
12.	COMPARISON OF IN-STACK AND IN-PLUME AVERAGE EMISSIONS IN TONNES/DAY	59
13.	EMISSIONS OF METALS FROM THE IRON ORE RECOVERY AND NICKEL REFINERY PLANTS IN KILOGRAMS/DAY	60
14.	AVERAGE LOW LEVEL EMISSIONS OF METALS FROM THE INCO SMELTER AT COPPER CLIFF DURING TYPICAL PRODUCTION RATES OF NICKEL BESSEMER MATTE AND BLISTER COPPER IN KILOGRAMS/DAY	62
15.	EMISSIONS OF METALS FROM THE FALCONBRIDGE 93 M STACK IN KILOGRAMS/DAY	64
16.	ACID EMISSIONS FROM MAJOR INDUSTRIAL SUDBURY SOURCES IN TONNES/HOUR	66

LIST OF FIGURES

	TITLE	PAGE
1.	LOCATION OF MAJOR AIR POLLUTION SOURCES IN THE SUDBURY BASIN IN ONTARIO	3
2.	SULPHUR DIOXIDE EMISSIONS COMPUTED BY MASS BALANCE VS EMISSIONS MEASURED AT THE INCO 381 M STACK	24
3.	SO_2 EMISSIONS VS ROASTER MATERIAL INPUT	26
4.	SO_2 EMISSIONS VS CONVERTER MATTE INPUT	27
5.	TOTAL PARTICULATE VS SULPHUR DIOXIDE EMISSIONS	37
6.	FREQUENCY DISTRIBUTION OF ERRORS FOR PARTICULATE VS SULPHUR DIOXIDE EMISSIONS	38
7.	IRON VS SULPHUR DIOXIDE EMISSIONS	51
8.	FREQUENCY DISTRIBUTION OF ERRORS FOR IRON VS SULPHUR DIOXIDE EMISSIONS	52
9.	COPPER VS SULPHUR DIOXIDE EMISSIONS	53
10.	FREQUENCY DISTRIBUTION OF ERRORS FOR COPPER VS SULPHUR DIOXIDE EMISSIONS	54
11.	NICKEL VS SULPUR DIOXIDE EMISSIONS	55
12.	FREQUENCY DISTRIBUTION OF ERRORS FOR NICKEL VS SULPHUR DIOXIDE EMISSIONS	56

ACKNOWLEDGEMENTS

This report is based on results published in many reports, and appreciation is extended to all individuals whose efforts have made this report possible.

Personnel from various process technology groups and laboratories of the Inco Metals Company and Falconbridge Limited, and Sudbury Regional Office, Laboratory Services Branch and Air Resources Branch staff of the Ministry of the Environment contributed to the generation of the data compiled and discussed herein.

Special thanks are extended to Messrs. G. Wong and J. Marson for performing calculations for this report, Messrs. R. Potvin, L. Fitz, E.T. Barrow, C.B. Martin, Drs. M. Lusis and W. Chan for their critical review of this report.

1. INTRODUCTION

The Sudbury Basin in Ontario is an area of major environmental interest because of large emissions of sulphur dioxide and particulates due to smelting and mining activities. In 1973, the Ontario Ministry of the Environment recognized the need for an evaluation of the impact of the smelting industries on the area and initiated studies aimed at characterization of emissions, in terms of both chemical composition and quantities, from the most significant sources in the Basin. In 1977, a more comprehensive program was established at the Air Resources Branch with the following objectives:

1. to define the sources and source strengths of atmospheric emissions in the Sudbury Basin.
2. to determine the fate of these emissions.
3. to determine the ambient air concentrations of potentially hazardous substances.

In essence, the study problem was the origin and fate of the pollutants, the nature and quantity of the emissions, and then dispersion, chemical transformation and ultimate deposition subsequent to being released to the atmosphere. A more detailed description of the program is given in the Ministry report ARB-27-82 ARSP (39).

This report deals with the emissions. The major emission sources in the Basin are described in detail and the emissions of pollutants are quantified on the basis of many reports issued in the last ten years by the companies which own the processing plants in the area and the Ministry of the Environment.

The processing plants with the major air pollution sources in the Sudbury Basin are all located within a distance of 15 kilometres from the city of Sudbury.

(the location of these plants is illustrated in Figure 1). They are associated with two copper and nickel smelters (Inco Metals Company at Copper Cliff and Falconbridge Limited in the town of Falconbridge) and the Inco Metals Company nickel refinery - iron ore recovery plant complex at Copper Cliff. Emissions from these sources account for almost all industrial emissions in the Basin.

During the eight years from 1973 to 1980, emissions from a number of individual sources at both smelters were measured by the companies and governments, and the combined emissions from these sources have been considered as representative of all industrial emissions in the Basin. Most of the emission measurements were done in stacks or ducts venting plant emissions to these stacks. In the case of Inco, emissions escaping from the main smelter building through windows and building ventilators were also measured.

A variety of measurement methods suited to a particular source or stack were employed by various industry or government groups. These methods ranged from rigorous source testing procedures as specified by the Ministry of the Environment to unproven and experimental emission measurement techniques. The observed inconsistencies and differences in the measured results were partially attributed to the nonrepresentativeness of some measurement methods.

Besides differences in the measurement methods applied at the same source at various times, there were differences in the process conditions, and particularly in the amount of materials processed in the measured plants. The variations in the process conditions effected variations in the emission rates and the results of the emission measurements. However, due to complexity of the processes involved in smelting, there were difficulties in relating the effects of a specific process (or changes in the operation of this process) on the total

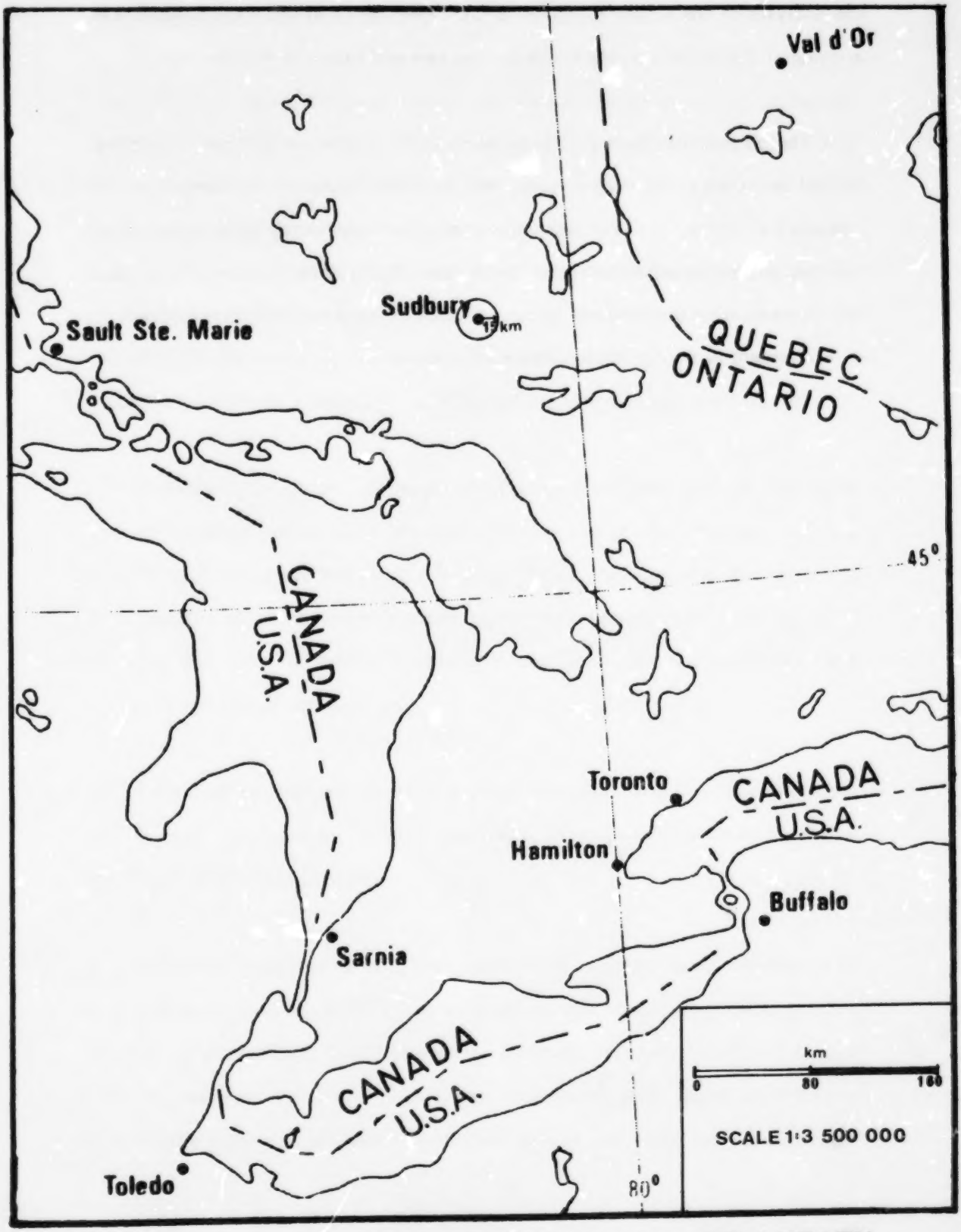

FIGURE 1. LOCATION OF MAJOR AIR POLLUTION SOURCES IN THE SUDBURY BASIN IN ONTARIO

emissions from the smelter or the plant. This in turn resulted in difficulties in the analyses of the results measured at the same source at different times and in arriving at the emissions rate which is most representative of the process.

The purpose of this report is to summarize and discuss all emission testing carried out during the period from 1973 to 1980, interpret and correlate the measured emissions with production wherever possible, and provide a technical basis for estimation of emissions in the Sudbury Basin. This in turn is necessary for the evaluation and analysis of air pollution in the Basin and consideration of any abatement action by the industries of concern.

2. SUMMARY

The most significant source of air pollution in the Sudbury Basin is the 381 metre stack at the Inco Copper Cliff smelter. Depending on the pollutant, this source may contribute anywhere from 50% to over 80% of all emissions in the Basin. The only exception is copper, which is also emitted in significant quantities from other sources. In order of emission magnitude the other sources of significant pollution in the area are the Inco 194 metre stack, the Falconbridge 93 metre stack, and finally ventilators and windows which emit unconfined emissions from Inco's main smelter building at Copper Cliff.

Emissions of almost all major pollutants from the Inco 381 m stack increase proportionately with production activities in the smelter. It was possible to demonstrate this relationship for this source because of the relatively large number of measurements performed in the period from 1973 to 1980. Corresponding data for the other sources in the Basin were not sufficient for a similar experimental verification.

Emissions of selected major pollutants from the measured sources in the Sudbury area are shown in Table 1. Maximum, average and minimum emissions are shown wherever available.

Emissions of sulphur dioxide from the Inco 381 m stack have been measured on a continuous basis since 1973, and were shown to change proportionately with the level of production. The emissions of particulates, iron, nickel, and copper from this stack as shown in Table 1 were calculated from linear correlations between the measured emissions of sulphur dioxide and these species such that

TABLE 1: YEARLY EMISSIONS OF MAJOR POLLUTANTS IN THE SUDBURY BASIN IN TONNES FOR THE PERIOD 1973 - 1981

Source	Variation	Sulphur dioxide [o]	Sulphuric acid	Total particulate	Iron	Copper	Nickel	Lead	Arsenic
Inco 381 m Stack*	Maximum	1,185,449	14,541	14,494	1,454	350	342	298	201
	Average	885,667	7,270	11,417	990	245	228	184	114
	Minimum	383,000	3,241	6,491	201	70	53	88	70
Inco 194 m Stack	Maximum	104,390							
	Average	54,568	1,664	2,380	643	171	226	6	4
	Minimum	18,980							
Inco Smelter (Low Level)	Maximum	16,000		755	90	312	40	0.8	0.1
	Average	12,000	88	586	70	242	31	0.6	0.1
	Minimum	6,000		283	34	117	15	0.3	0.1
Falconbridge 191 m Stack	Maximum	274,000							
	Average	173,000	438	865	98	11	9.6	13.4	6.4
	Minimum	88,000							
Inco** Two 45 m Stack	Average	-	-	(4,073)	(2,354)	-	-	-	-
TOTAL	Average	1,125,235	9,460	15,248	1,801	669	500	204	125

Basis: 365 days x 24 hrs/day production.

* Oxides of nitrogen emissions expressed as NO_2 = 3,281 tonnes.

* Hydrogen chloride emissions = 530 tonnes.

* Emissions of particulates, iron, copper and nickel from the 381 m stack are yearly values calculated from correlations with sulphur dioxide emissions. Emissions of sulphuric acid, lead and arsenic are average measured values.

** Emissions ceased in April 1980, not included in average.

o Except for INCO Smelter (low level), all sulphur dioxide emissions were obtained by mass balance calculations. Annual low level ammonia emissions from Inco's iron ore recovery plant in 1977 was 4,219 tonnes.

they correspond to annual sulphur dioxide emissions. In this way, the effect of production rates on emissions has been taken into account, and so the calculated emissions as shown in Tables 1 and 10 are considered more representative of the actual emissions than the individual emissions measured in stack tests of short duration shown in Table 11. Before selecting the straight-line correlations, a number of linear and nonlinear relations were examined for the goodness of fit through the experimental points plotted on the graphs. In the case of iron, copper and nickel, the straight lines and exponential curves provided reasonable correlations of the data available; however, since the estimates from these functions were not significantly different, it was decided to use the mathematically simpler straight-line correlations. As is the case with any function fitted through scattered experimental points, errors (or uncertainties) are associated with the estimates of emissions of particulates, iron, copper and nickel, calculated from the straight-line correlations in this report and these errors must be taken into account when using the calculated emissions. In any individual case, the variation from the straight-line estimate can be greater than $\pm$100%, but is generally less than $\pm$75%. The indicated maximum, average, and minimum in Table 1 are yearly values. Emissions during shorter periods in any of the examined eight years could be either smaller or larger, depending on production. Emissions from the other sources in Table 1 were measured in shorter periods and are in some cases based on the results of one test only.

About 78% of all sulphur dioxide and 77% of all sulphuric acid emissions are from the Inco 381 m stack; the estimates of total particulates indicate that the corresponding contribution from this source is lower at 75%, while the contributions to metal emissions are still smaller at 37-55% except for lead, and arsenic which are mostly emitted from this source.

Up to April 1980, when the pelletizing plant, associated with the iron ore

recovery process was shut down, Inco's two 45 m stacks at this plant were significant sources of particulate and iron emissions in the Basin. The best available emission data are based on a single testing survey of only one stack. Hence, due to the paucity of data for this source, the emission rates presented in Table 1 were not included in the average emissions from the Sudbury Basin. This source is not expected to resume operation.

Emissions of oxides of nitrogen and hydrogen chloride were measured on two occasions at the Inco 381 m stack, and because they were small in comparison to sulphur dioxide and sulphuric acid, these measurements were discontinued at this and other stacks in the Basin.

Ammonia emissions are low level emissions, occurring through losses to the atmosphere via leaching and recovery buildings' ventilators. They are calculated from losses in process and discharge streams on a monthly basis. Annual emission for 1977 was the only available estimate at the time of writing.

The size of the emitted particulates is an important consideration in the understanding the deposition of particulate pollutants and in determining the airborne part of particulate emissions. Since most sources in the Sudbury Basin are controlled and the majority of particulates are removed by electrostatic precipitators at a source, it was expected and confirmed by testing that large portions of the particulates in emissions are less than 10 micrometers (m μ) in diameter.

3. DESCRIPTION OF SOURCES AND PROCESSES

3.1 GENERAL

The most significant sources of industrial emissions in the Sudbury Basin are five stacks - the Inco 381 m and 194 m stacks, the Falconbridge 93 m, and prior to 1980, Inco's two 45 m stacks. Before entering these stacks, emissions from various industrial processes are cleaned of a large portion of particulate pollutants by electrostatic precipitation and contain small and to a large extent airborne particulates. These are called confined emissions, which after being discharged are mostly carried away from stacks and are diluted considerably with ambient air before impinging on the ground. Other emissions are unconfined, since they escape various process fume collection steps in the ore and metal processing. They are emitted through small stacks on building roofs, windows and ventilators, and called low level emissions, impacting in the plant vicinity and reaching the ground in a more concentrated form. They are difficult to quantify and were measured by the company at Inco's Copper Cliff smelter building only. However, this building is the most significant source of low level emissions in the Sudbury Basin.

A detailed description of the processes described in this chapter was presented in an earlier Ministry report (24). The following is intended as a summary of that information.

3.2 INCO'S 381 M STACK AND THE SMELTER BUILDING AT COPPER CLIFF

The 381 m stack emits gases from pyrometallurgical smelting processes at Inco's Copper Cliff smelter. The outside diameters of this stack at the base and

at the top are 35.5 m and 15.8 m, respectively. About 157,000 cubic meters of gas at a temperature of 130°C are emitted each minute from this stack. The gas temperature increases with increasing production rates and can reach about 175°C when the smelter is operated at full capacity. Gases are received from various processes via two inlet breachings situated opposite to each other at the stack base, and are cleaned of most of the particulate pollutants by electrostatic precipitators called Cottrells prior to entering the stacks.

Because of the large stack diameter, the positions of the two inlet breachings and the location of the existing sampling platform, strict emission measurements at this stack are long lasting, manpower intensive and expensive. On occasions, the measurement methods were simplified at a cost of somewhat reduced accuracy.

The purpose of the smelting processes which emit pollution into this stack is to make nickel and copper of appropriate purity in the form of nickel bessemer matte and blister copper from suitably prepared ore concentrates. There are two main circuits with associated process equipment - one of each for nickel and copper. Nickel matte processing is considered a separate process, however, since in comparison to ore roasting and smelting, it is not a major source of emissions, this process will not be discussed any further.

3.2.1. COPPER CIRCUIT

Copper concentrate, mixed with sand, is smelted in a flash furnace, producing copper matte, slag , and sulphur dioxide at about 80% concentration in the off-gases. These gases are first cleaned in scrubbers and a wet electrostatic precipitator, then sent to the sulphur products plant for the production of liquid sulphur dioxide. In case of an emergency, these gases may be directed to the 381 m stack via a bypass system. The gases rising from the handling operations of

the two melted products - slag and copper matte - are split, with the major portion being emitted via a small stack on the roof of the building and the rest directed to the 381 m stack. In addition to these major emissions, there are fugitive emissions resulting from leakages in the scrubbers, sulphur dioxide release from the scrubber liquid, temporary pressure increases in the furnace and the periodic return of the converter slag.

Copper matte from this furnace is charged to copper converters to eliminate the remaining iron and to produce blister copper which is then sent to copper refinery for further refining. Converting is a batch process in which oxygen enriched air is blown through the melted charge to produce oxides of iron which react with silica and form slag. The main product of converting, blister copper, is sent to the copper refinery for further processing. The main portion of off-gases is sent to the 381 m stack via collection fume hoods and flues. Part of the off-gases escapes from around the collecting hoods into the building and outside through the roof ventilators as fugitive emissions.

3.2.2 NICKEL CIRCUIT

Nickel concentrate is first mixed with sand and then roasted in multi-hearth roasters to remove moisture and a part of sulphur as sulphur dioxide. Heating of concentrate on the roaster hearths is accomplished by the combustion of natural gas and the oxidation of iron sulphide with air. This air along with sulphur dioxide and entrained particulates is directed to the 381 m stack via piping arrangements on the roasters' tops. Besides these confined emissions, there are periodic fugitive emissions escaping through the inspection doors. These emissions rise to and are emitted through the roof ventilators and the building windows. Six roasters are operated in conjunction with one reverberatory furnace.

The product of roasting, calcine, is fed to the reverberatory furnaces to combine iron oxide with silicate and produce slag. Slag floats on the top of matte which contains heavier sulphides of copper, nickel and iron. Combustion of oil, sustained with oxygen enriched air, provides the necessary heat to maintain the furnace contents in a molten state. The combustion gases with entrained particulates and sulphur dioxide are conveyed via a reverb-header flue to the 381 m stack. Unconfined fugitive emissions from this furnace originate during the removal of matte and slag from the furnace. They are collected by hoods and exhausted into separate vents. In addition, puffing emissions, escaping the furnace through its brickwork during periods of increased pressure within the furnace, and the emissions originating during the converter slag removal exit the smelter building via the roof ventilators above the furnace. There are five reverberatory furnaces of which three or four are usually operated at any one time.

Matte from these furnaces is fed to the nickel converters, where similar operations to those in copper converting are carried out. After oxidation of iron and formation of slag, the converter is rotated for skimming and discharging of a high grade copper-nickel matte for casting and cooling. Emissions from various steps - charging, blowing, skimming and pouring - are collected by hoods and directed to the electrostatic precipitators and the 381 m stack. However, some of these emissions escape around the hoods and from the building via ventilator openings. Fourteen converters are normally used in the nickel circuit.

3.3 THE INCO 194 M STACK AND THE IRON ORE RECOVERY PLANT COMPLEX AT COPPER CLIFF

The Inco 194 m stack is the second largest source of industrial emissions in

the Sudbury Basin. Approximately 9500 cubic meters of gas at 150°C to 200°C, containing metals, sulphur dioxide, and sulphuric acid are exhausted every minute from this stack. The stack receives off-gases from Inco's iron ore recovery plant, nickel refinery and three sulphuric acid plants. Similarly to process off-gas treatment at the nickel smelter, emissions from these five plants are cleaned of the majority of entrained particulates prior to entering the stack. Cyclones, scrubbers, settling chambers, electrostatic precipitators and combustion units are used for this purpose at the iron ore recovery plant, electrostatic precipitators at the nickel refinery, and packed water scrubbers at the three acid plants.

Emissions from the 194 m stack were not measured directly in the stack, but in the flues from the iron ore recovery, the nickel refinery and the acid plants. These flues were not readily amenable to stack sampling and the measurement methods had to be compromised and less rigorous procedures used because of limitations in locating suitable sampling sites. Consequently, the accuracy of measurements was affected and somewhat larger errors are associated with the reported emissions from the 194 m stack.

3.3.1 IRON ORE RECOVERY AND PELLETIZER PLANTS

There are four processes involved in iron ore recovery at the Inco plant: roasting and reduction of pyrrhotite concentrate, leaching, recovery and pelletizing. The purpose of these processes is to recover nickel from pyrrhotite concentrate received from the Inco smelter and to prepare iron rich pellets for marketing. In the roasting step, most of the sulphur is eliminated by oxidation at high temperatures. Off-gases from six fluid bed roasters are cleaned of particulate materials and sent to the acid plants. These gases can be temporarily diverted to the 194 m stack in case of upsets, however, if problems persist, the plant is shut down. To facilitate the separation of nickel from iron

in the subsequent leaching and reduction steps and to reduce the oxides of iron, the concentrate from the roasters is treated in six reduction kilns. Off-gases from six fluid bed roasters are cleaned of particulate materials and sent to the 194 m stack. It can be suggested on the basis of stack tests that the reduction kilns are a major source of particulates emitted from the 194 m stack. Leaching and recovery plants are sources of ammonia emissions from small stacks on the building roof or process equipment vents -ammonia is used in the process of nickel recovery from the reduced concentrate. The concentrate, removed of nickel, copper and cobalt, is sent to the iron ore pelletizing plant to produce concentrate pellets rich in iron and suitable for marketing. In order to produce the pellets, the leached magnetite from the leaching plant is first dewatered to make filter cake, which is then dryed in two sintering machines. Gases from these machines are emitted via two 45 m stacks and four small stacks on the building roof. The two 45 m stacks were major sources of iron and total particulate emissions in the Basin until the plant shut down in April, 1980.

3.3.2 NICKEL REFINERY

Nickel sulphide from matte processing is further processed in the nickel refinery to make nickel in either powder or pellet forms. Nickel sulphide, precious metal intermediate products, and refinery residues are first briquetted and then oxidized in two converters. The finished metal is further processed to produce metallic granules which undergo carbonylation, purification, decomposition and either pelletization or powderization. The most significant source of emissions to the 194 m stack is the converting step. The gases exhausted from the converters are passed through electrostatic precipitators prior to entering the flue leading to the stack. Even though converting is a batch process operated intermittently, these two converters were the second largest

source of nickel emissions in the Sudbury Basin up until 1980, when additional precipitation capacity on each converter was installed.

3.3.3 SULPHURIC ACID PLANTS

Inco also operates three sulphuric acid plants, adjacent to the iron ore recovery process from which they receive sulphur dioxide. The sulphuric acid process involves oxidation of sulphur dioxide on a catalyst to produce sulphur trioxide, and absorption of sulphur trioxide in absorption towers. Residual sulphur dioxide gas not converted to acid (conversion efficiency of about 93%) is vented to the atmosphere via the 194 m stack.

3.4 THE FALCONBRIDGE 93 M STACK

About 7100 standard cubic meters of gas at 120^{o} - $160^{o}C$ are emitted from this stack every minute. The stack receives off-gases from the Falconbridge smelter operations, which include smelting and converting, as well as the off-gases from the adjacent sulphuric acid plant. Prior to entering the stack, the process gases pass through an electrostatic precipitator for the removal of entrained particulates.

The Falconbridge smelting process was changed in 1978, hence, the emissions from the plant should be considered in two periods: before and after this change. There were no satisfactory emission measurements performed on the old process and the only acceptable emission estimates up to 1978 are those of sulphur dioxide as calculated by mass balance. Emissions of some metals from the old process were also measured on two occasions, however, the accuracy of these measurements was judged unsatisfactory. Since 1978, two lines have been installed at the smelter, each containing a fluid bed roaster and

an electric furnace, replacing the sintering machines and blast furnaces in the old process. The off-gases from the roasters are cleaned of particulates in cyclones and electrostatic precipitators, cooled, and sent to an acid plant for the conversion of sulphur dioxide to sulphuric acid. The solids from the roasters are fed to electric furnaces for smelting. Similarly to the roaster off-gases, emissions from the furnaces are first passed through cyclones and electrostatic precipitators and then sent to the stack. The furnace matte is sent to converters for further removal of iron and sulphur. There are four converters of which three are operated at any one time. The off-gases from the converters are sent to the electrostatic precipitators and then to the stack.

4. DISCUSSION OF EMISSION MEASUREMENT METHODS, THEIR FREQUENCY AND ACCURACY

The emission measurement methods applied at the Sudbury sources varied in complexity and accuracy, depending on the source and the manpower available for these measurements. They were, in most cases, either identical to or modified methods in the Ontario Source Testing Code (OSTC) (36). When modifications to the prescribed methods were made, it was either demonstrated that the measurement accuracy was not affected to a large degree or the construction limitations at the given sources were such that no other acceptable alternative existed at the time. In some cases, novel and unconventional methods, such as in-plume measurements, were applied. At most sources, measurements were done in one period of short duration and, consequently, the results reflect the emissions in effect during that period.

At the largest Sudbury source, the Inco 381 m stack, the particulate emission measurements were done at least once a year in the period from 1973 to 1980, producing relatively abundant emission data points. These emission data were correlated with the emissions of sulphur dioxide, thus enabling prediction of particulate emissions, within the given error margin, at any time, providing that either the production rates in the smelter or the emissions of sulphur dioxide from this stack are known. Emissions of sulphur dioxide from this stack are measured continuously with a continuous emission monitoring system, however, in the absence of measured values, these emissions could be calculated by mass balance. The continuous emission monitoring system at the 381 m stack was exhaustively examined and calibrated against a reference method in 1973, and it was then demonstrated that the system was accurate to within 10%.

Measurements of particulate pollutants at the 381 m stack in 1973 and 1974 were done according to rigorous testing procedures, and the obtained results are believed to be the most accurate of all the emission data available for this stack. In 1974, attempts were made to simplify testing procedures and special tests to compare so called one-point sampling with multi-point sampling techniques were carried out for the first time. Starting in 1975 only one-point sampling had been used at this source until 1980, when another simultaneous single-point and multi-point testing program was performed. These tests were designed to measure the magnitude of error associated with single-point sampling. The results suggested that single-point sampling could underestimate emissions by as much as 60% (22). A similar magnitude of error was indicated from the correlations between the emissions of particulates and sulphur dioxide from this stack, as discussed later in this report.

Besides errors arising from single-point sampling, there were inconsistencies in the chemical treatment of particulate samples in different years, causing difficulties in comparing the measured yearly values. In order to remove these difficulties and, at the same time, to conform to the definition of particulate matter as given in the OSTC, sulphates and free sulphuric acid found prior to and on the filters of the sampling trains after the tests were considered particulates and were included in the calculation of particulate emissions. This still did not eliminate all inconsistencies, particularly those due to different distribution of acid in the sampling trains caused by different filter temperatures during samplings, but it helped to obtain better correlations between the yearly results.

Measurements of sulphuric acid emissions from most of the Sudbury sources are difficult due to limitations of the sampling method. The method used in the measurements is known as the U.S. E.P.A. Method 6 (37), which is

directly applicable to sulphuric acid plants, but had to be modified for the other sources in Sudbury. These modifications were not always successful, and difficulties were met when trying to separate sulphates from free acid during sampling and to distinguish between these two species in subsequent chemical analyses. Selective chemical extractions and elevated temperatures of the probes and filters of the sampling trains during sampling applied in latter years helped, but did not completely resolve the problem. In some tests, the solvent used for the extractions was not completely selective because of impurities, and the sampling temperatures were not maintained sufficiently high because of sampling equipment limitations.

Sulphur dioxide is another possible interference in the acid emission measurements. It can be either oxidized under favourable conditions to form sulphuric acid or retained in the impinger solutions to interfere in the chemical analysis. While both of these mechanisms can be controlled by careful preparation and execution of sampling, it is possible that in the actual measurements this was not always achieved, and that the sulphuric acid results from some tests are biased.

Emission measurements of most trace metals were more accurate than either particulate or acid measurements. Error in these measurements were due to single-point sampling and the chemical analyses of the samples. The only exceptions are volatile metals such as mercury, arsenic, and selenium, which were determined accurately on one occasion at the 381 m stack by using a specially designed emission measurement method in 1980.

Emission measurements at the Falconbridge stack and the Inco 194 m stack were also difficult due to problems similar to sampling at the 381 m stack: traversing of stacks and flues was incomplete due to practical limitations and

samples of particulates and sulphuric acid were subject to similar interferences. These problems are fully described in the discussions of emissions of individual species from these sources.

Mass balance was used to estimate sulphur dioxide emissions, whenever possible, and these estimates can be considered accurate to within 10%, as will be discussed later.

Ammonia emissions from Inco's iron ore recovery plant were estimated by mass balance, based on measurements related to chemical makeup, liquid effluent loss, and specific process emission vent losses, with the remaining unaccounted losses included as part of atmospheric emission loss (24).

The low level emissions from the Inco smelter building as given in the original testing reports prepared by the company (12, 14, 17, 20, 21) are probably the least accurate of the emission data presented in this report. This was due to difficulties in sampling, which are typical in fugitive emission measurements, for the following reasons: emissions are intermittent in both strength and duration; sampling areas are large, with uneven gas flow and occassionally low gas velocities; problems are experienced in locating "representative sampling points"; there are difficulties in assigning a portion of unconfined and wandering emissions to a specific process unit within the building; and, commercially available sampling equipment is not suitable for these measurements and special sampling trains have to be built. Low level emission measurements at Inco's smelter were done on a process unit basis, where a reverberatory furnace, a nickel converter, a multiple hearth roaster, the copper flash furnace and a copper converter were selected as representative of other corresponding process units in the building, and the measured emissions prorated to include all process

units. Measurement errors associated with individual process units are given in the testing reports and they were used to calculate the total error in the reported low level sulphur dioxide emissions from the smelter. This error was ±27% (33). The error in particulate emissions was not estimated, but it is expected to be larger than in the case of sulphur dioxide because of possible stratification effects on sampling accuracy.

The size of emitted particulates from the stacks was measured by impaction techniques and generally these measurements are considered accurate. When there were doubts about the execution of some tests, the tests were repeated and unreliable results discarded.

In-plume measurements were also attempted as a substitute for in-stack emission measurements. Sometimes the results from the two methods were fairly similar, however, on occasion the differences were large and impossible to explain. In these cases it was assumed that the in-stack method was more accurate.

5. SULPHUR DIOXIDE EMISSIONS

Yearly sulphur dioxide emissions from most smelting processes could be conveniently calculated by a mass balance procedure. However, difficulties and inaccuracies could arise, if the emission data from more than one stack which receive off-gases from the same process are required. In these situations, sulphur dioxide measurements at each stack is the only viable alternative. Measurements will also produce more accurate emissions over shorter periods, e.g. days or weeks, when inaccuracies in process sampling and weighing of the process streams required for mass balance could have a more significant effect on the accuracy of emission calculations.

A combination of measurements and mass balance was used to produce sulphur dioxide emissions from the major industrial sources in the Sudbury Basin as shown in Tables 1 and 2. Emissions from the Inco 381 m stack are mass balance values as reported by the company and they agree with the yearly average of the measured values to within 1-2%. Since the measured values were shown to be accurate to within 10% (1), it is likely that the mass balance values in Table 2 are at least as accurate. As far as other emissions in Table 2 are concerned, the Falconbridge and Inco 194 m stack data were obtained by mass balance, while the Inco smelter low level emissions were measured in 1978 and 1979, and prorated to other years on the basis of production.

For Inco, shutdowns and/or labour strikes in 1978 and 1979 were responsible for lower emissions in the Basin in those years. Sulphur dioxide emissions for 1980 and 1981 from the Inco 381 m stack reflect more stringent emission requirements imposed by the Ministry in 1980 which were met as a result of reduced production. With respect to emissions from the Falconbridge smelter, the levels were reduced as of 1978 largely due to process changes (installation of

fluid bed roasting - electric furnace smelting - sulphuric acid manufacturing). However, summer vacation shutdowns and lower production levels also contributed to the lower emissions presented in Table 2. In other respects, the tabulated emissions should be regarded as representative of each source. The measurement methods, correspondence between emissions and production as well as the accuracy of prorated emissions given in Table 2 are discussed for each source separately.

TABLE 2: YEARLY EMISSIONS OF SULPHUR DIOXIDE FROM MAJOR INDUSTRIAL SOURCES IN THE SUDBURY BASIN IN THOUSAND TONNES

Year	Inco 381 m Stack*	Inco Low Level Smelter**	Inco 194 m Stack*	Falconbridge 93 m Stack*
1973	1,171	14	104	274
1974	1,126	16	74	258
1975	1,141	16	40	195
1976	1,163	15	41	191
1977	1,069	15	52(55***)	200
1978	535	8	24	117
1979	383	6	19	88
1980	733	11	66	123
1981	650	9	64	114
Average	885	12	54	173
% of total	78	1	5	16

* Determined by mass balance calculations
** Determined from measurements in 1978 and 1979 and prorated to other years on the basis of production.
*** Determined from stack measurements.

5.1 THE INCO 381 M STACK

With 78% contribution to the total emissions of sulphur dioxide in the Sudbury Basin, the 381 m stack is the largest single emitter of sulphur dioxide in the area. The accuracy of total sulphur dioxide emissions from the Basin depends to a large extent on the accuracy of these 381 m stack emissions, and it is for this reason that they have been measured on a continuous basis since 1973.

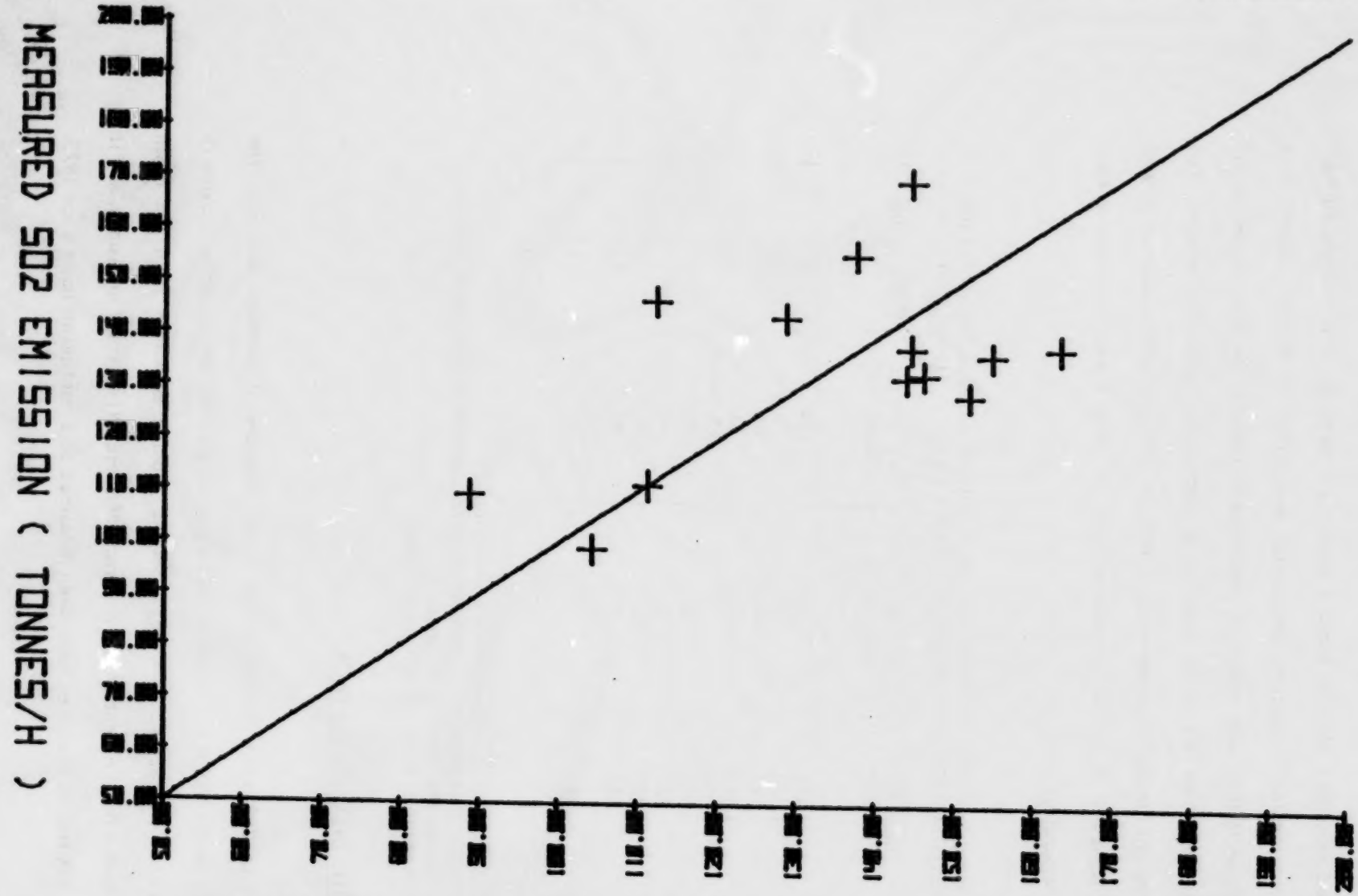

FIGURE 2. SULPHUR DIOXIDE EMISSIONS COMPUTED BY MASS BALANCE VS EMISSIONS MEASURED AT THE INCO 381 M STACK

The continuous emission monitoring installation for sulphur dioxide was verified through extensive testing in 1973, when it was demonstrated that the system was accurate to within ± 10%. The measured emissions were also compared with the available computational estimates by mass balance in 1973 and 1974, and a relationship between these two sets of results is shown in Figure 2.

The plotted line in Figure 2 represents an ideal one-to-one relationship, assuming no error by either method. The measured values are spread around this line with the largest individual point deviation being about 30%, which, considering that both the continuous monitoring and short term mass balance methods are susceptible to errors, should not be unexpected. Figure 2 still illustrates that the two methods are essentially equivalent over longer time periods. This equivalency has also been confirmed in Inco's progress reports on continuous monitoring, which indicate that yearly values obtained by the two methods agree to within 2%, while monthly averages may differ by as much as 16% (32).

The available measured emissions were also plotted against the total ore concentrate input to the roasters and the total feed to the nickel converters in Figures 3 and 4, respectively. Straight best fit lines shown in these figures were calculated by the least squares method.

The large scatter of points about the lines in Figures 3 and 4 may be a result of the variable sulphur content of the feeds to both the roasting and converting operations. More uniform sulphur content of the reverb matte fed to the converters resulted in better correlation and smaller scatter in Figure 4. From these figures, it is calculated that, depending on feed rate, sulphur dioxide emissions range from 0.66 to 0.78 tonnes for every tonne of the total material input into the converters and from 0.61 to 0.84 tonnes for every tonne of the ore concentrate fed into the roasters.

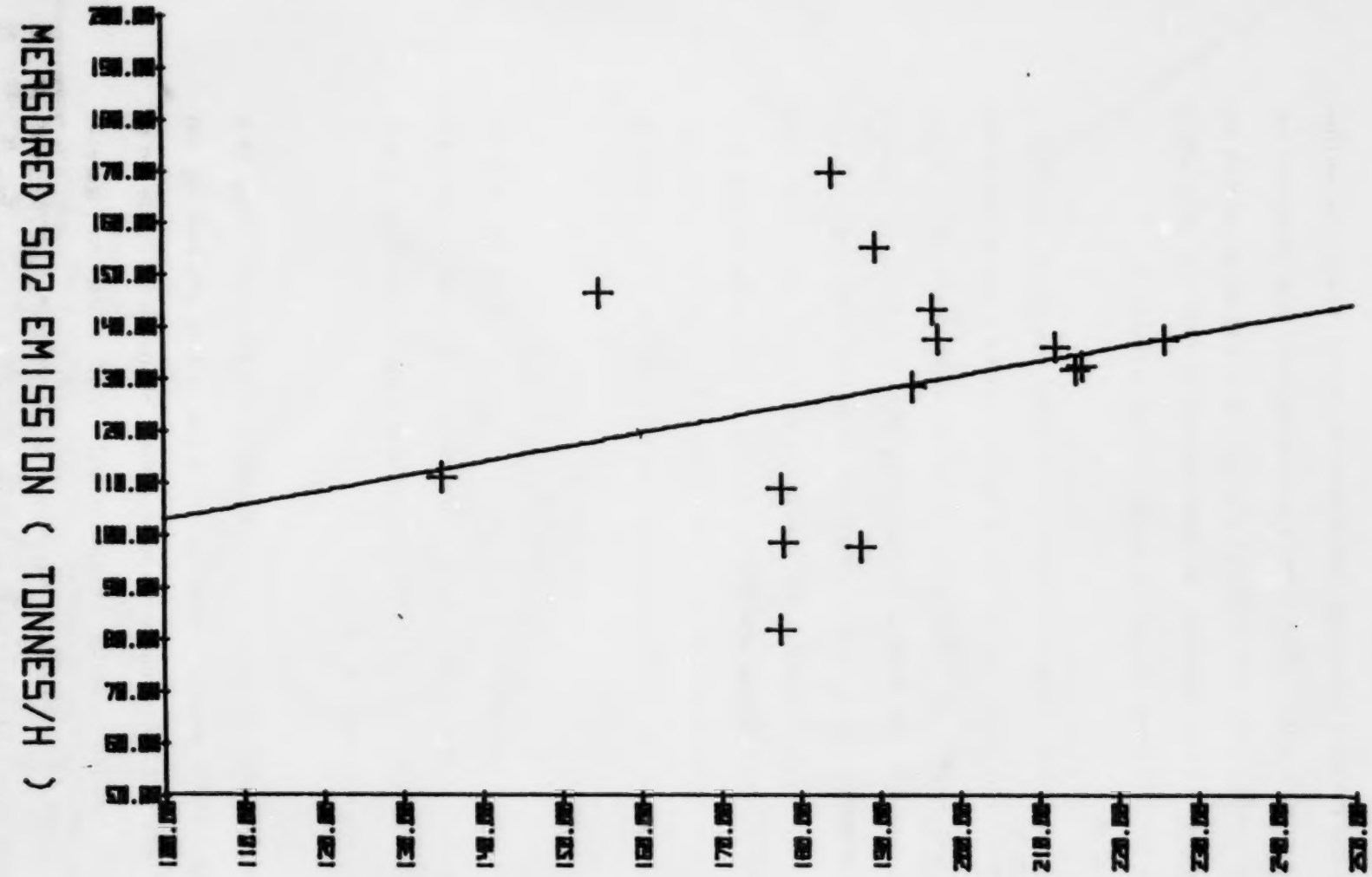

FIGURE 3. SULPHUR DIOXIDE EMISSIONS VS ROASTER MATERIAL INPUT

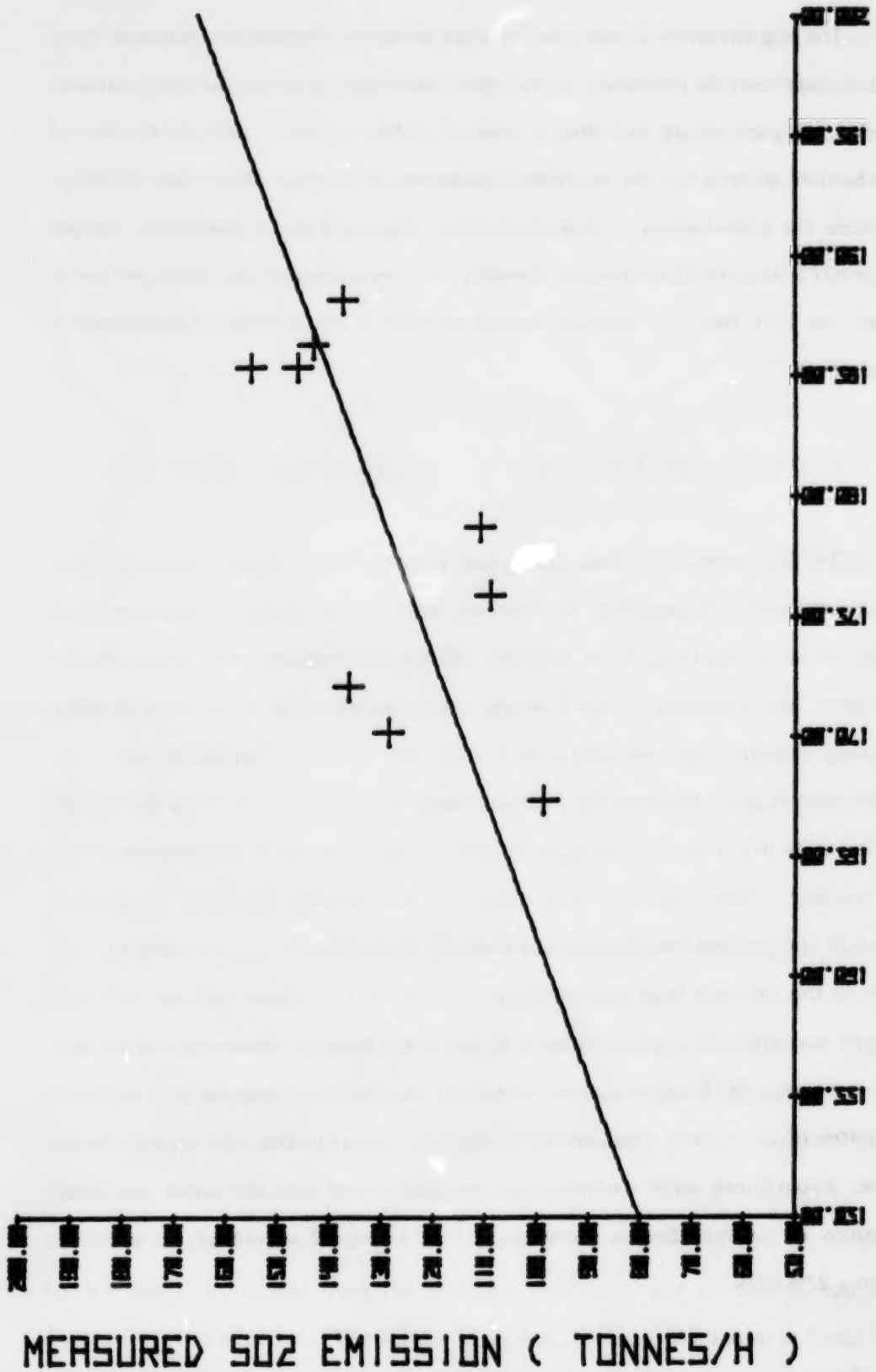

FIGURE 4. SULPHUR DIOXIDE EMISSIONS VS CONVERTER MATTE INPUT

The significance of the relationships between the process material flows and sulphur dioxide emissions is that they illustrate a proportionality between these two parameters and enable use of either of them as an indicator of production activity in the smelter. However, a caution should be exercised because the correlations as calculated from Figures 3 and 4 may have changed since 1973, as a result of process changes, but these changes are not expected to alter the fact that the sulphur dioxide emissions are directly proportional to production.

5.2 INCO LOW LEVEL EMISSIONS

The low level emissions from the Copper Cliff smelter building were measured by Inco personnel in a series of tests at various vents, windows, and small stacks on the roof of the building. No similar measurements were done at the other Inco buildings or the Falconbridge smelter. The tests at Inco were specially designed and executed such that the measured emissions from the single process units selected for measurements as representative of a number of similar units in the same building were used to calculate the total emissions from the building. The origin of these emissions was already discussed in general terms in the process description; however, for more details, reference should be made to the relevant Inco reports (12, 14, 17, 20, 21). Emissions shown in Table 2 were prorated to average yearly production rates of bessemer matte and blister copper. Difficulties encountered in these measurements as well as some estimates of associated measurement errors are given in the Inco reports listed above. From these error estimates, it was calculated that the total low level emissions of sulphur dioxide shown in Tables 1 and 2 should be accurate to within $\pm$ 27% (33).

From among the individual processes, the largest contributor to these low

level emissions of sulphur dioxide are the reverberatory furnaces which account for about 42% of the total low level sulphur dioxide emissions, followed by the converters at 38%, the copper flash furnace at 19% and the roasters at 1% (21). From another Inco report (27), the contributions to the total emissions for the copper converters are 41%, followed by the nickel reverberatory furnaces at 25%, the copper flash furnace at 17%, the nickel converters at 8% and miscellaneous sources at 9%.

5.3 THE INCO 194 METRE STACK

The flues from Inco's nickel refinery, the sulphur products plant and the iron ore recovery plant are joined to the 194 m stack. Because of the unavailability of suitable sampling platforms at the stack, all emission measurements were carried out at the three flues that convey the emissions from the individual plants to the stack. Sulphur dioxide emissions and the corresponding production rates, as given in the emission testing reports (8, 9, 10), are shown in Table 3.

Sulphur dioxide emissions in Table 3 are based on the results of several tests of limited duration carried out when the production rates were as indicated. Only in the case of the nickel refinery were the emissions prorated to correspond to a typical production rate in 1977. These nickel refinery emissions are intermittent, and the result in Table 3 corresponds to approximately three "converter heats" per day, each heat lasting about four and a half hours. The emissions during the heat changed from a low of 5% to a high of 60% of the total for the heat. Emissions from the sulphuric acid plant are continuous and also dependent on production. The sulphuric acid production rate shown in Table 3 is somewhat less than the maximum rated capacity of 2721 tonnes per day.

Emissions from the iron ore recovery plant are continuous and subject to fluctuations. The fluctuations were so large during testing that some doubts about the validity of tests and the meaning of the obtained results were expressed (29). Fortunately, this plant is the least significant contributor of sulphur dioxide emissions to the 194 m stack, and any error in these measurements will not affect significantly the accuracy of the total emissions from the stack.

All emissions from individual flues were measured instrumentally, and in the case of the nickel refinery, they were compared with mass balances calculated over both short and long time periods (9). The agreement between the measurements and long-period calculation was excellent, while short period mass balance indicated about 62% lower emissions.

TABLE 3: SULPHUR DIOXIDE EMISSIONS FROM INDIVIDUAL PROCESSES TO THE INCO 194 METRE STACK IN TONNES/DAY IN 1977.

Source	Sulphur Dioxide Emissions	Production Rate
Sulphuric Acid Plants	91.9	2300 tonnes/day of sulphuric acid
Nickel Refinery Plant	48.2	214.2 tonnes/day of dry charge to refinery
Iron Ore Recovery Plant	9.4	2267 tonnes/day of roaster feed
Total	149.5	

5.4 THE FALCONBRIDGE 93 M STACK

This stack is the second largest sulphur dioxide emitter in the Sudbury Basin, contributing on average about 16% of the total over the nine year period, as indicated in Table 2. These emissions have been computed by mass balance and reported by the company (13). In 1979, they were also measured over a short period, and the measured emissions were compared with the mass balance values. The comparison was not successful in that the measurements indicated about 49% higher emissions, probably as a result of errors in both the measurement and short term mass balance. However, the values in Table 3 should still be considered as an accurate representation of the emissions from this stack. The Falconbridge plant emissions are variable due to a batchwise operation of the converters and possibly variable efficiencies of the sulphuric acid plant, which both release off-gases to the 93 m stack.

6. EMISSIONS AND SIZE OF TOTAL PARTICULATES

Total particulates are the second most significant pollutant emitted from the industrial sources in the Sudbury Basin. However, in comparison to sulphur dioxide, the particulate emissions amount to only about 1.8% of the sulphur dioxide emissions.

Similarly to sulphur dioxide, most of the particulate emissions in the basin are due to Inco's 381 m stack. The 381 stack emissions, as shown in Tables 1 and 4, were obtained from a linear correlation between the particulate and sulphur dioxide emissions measured in the period from 1973 to 1980 such that they correspond to yearly emissions of sulphur dioxide. Since the sulphur dioxide emissions depend on the production rates in the smelter, the estimated particulate emissions are indirectly normalized with respect to production and represent the best available average particulate emissions from this source. Since about 75% of the total particulate emissions in the Sudbury Basin are due to the 381 m stack, any variation in these emissions will have a significant effect on the overall emissions of total particulates as well as of the other particulate pollutants in the Basin.

The low level particulate emissions as shown in Table 4 were also normalized with respect to production; however, in this case only one set of measured values from Inco's 1978-1979 testing program was available. Consequently, the calculated emissions shown in the table are less accurate then those for the 381 m stack. No such normalization was done for any other source listed in Table 4, and the indicated emissions from these sources were measured in only one set of tests. Emissions from the two 45 m smokestacks ceased in

April 1980, when the pellet plant was shut down (25). Emissions from the Inco 194 m stack have also been reduced since 1980, when additional precipitators were installed at the nickel refinery flues. Presently, no satisfactory stack test data are available for the estimation of this reduction.

TABLE 4: YEARLY EMISSIONS OF TOTAL PARTICULATES IN TONNES FROM MAJOR SOURCES IN THE SUDBURY BASIN FROM 1973 TO 1981.

Year	Inco 381 m Stack	Inco Smelter Low Level Emission	Inco 194 m and two 45 m Stacks	Falconbridge 93 m Stack
1973	12,166	650		
1974	14,144	730		
1975	14,299	755		
1976	14,494	721		
1977	13,763	703	2,380**	
1978	8,057	387		
1979	6,491	283	4,064*	865
1980	10,096	548		
1981	9,241	497		
Average	11,417	586		
Standard Error of Estimate	Sxy=4,588			

* Emissions from two 45 m stacks not applicable after April 1980, due to plant shutdown.

** These emissions were decreased in 1980 as a result of additional precipitator capacity at the nickel refinery plant flues.

6.1 THE INCO 381 M STACK

Emissions of total particulates from the 381 m stack were measured in at least one short period every year from 1973 - 1980. The average results of these measurements are shown in Table 5. They were based on: one test in 1975, two tests in each of 1976, 1978, and 1980, four tests in each of 1974 and 1977, six

tests in 1973 and 16 tests in 1979. Other tests that could have been done in any of these years were excluded because of a lack of detailed data on the chemical composition of the samples. Since the number of tests, the time periods of testing, and the production rates differed in each of these years, the emissions in Table 5 cannot be compared directly with each other or considered as representative of yearly emissions. Also, they are not identical to the emissions given in the original test reports because of the inclusion of the condensed acid in the particulate emissions. This inclusion was done to enable a direct comparison of emissions measured in different years and to conform with the definitions in the provincial testing code. In earlier reports, the original emissions included the part of sulphuric acid which was condensed at the sampling temperatures while the particulate emissions in later reports were completely free of this acid (different chemical treatments of particulate samples in different years were responsible for these inconsistencies). In the 1973 and 1974 tests, particulate samples were extracted with acetone which removed some of the acid, and these partially acid-free particulates were used to calculate the particulate emissions. Starting in 1975, isopropanol replaced acetone because of its effectiveness as a selective extractive chemical for the free acid and, consequently, the emissions after 1975 did not contain any free sulphuric acid. In order to make all emissions measured at this stack comparable, the condensed acid, which at the sampling temperatures had been collected as a particulate, was included in the particulate emissions. In doing so, these emissions also conformed with the definition of particulates given in the provincial testing code (OSTC). Additional benefit was a slightly better predictability of particulate emissions from the knowledge of production and emissions of sulphur dioxide.

The individual particulate emission results which were used to obtain the average emissions shown in Table 5 and the sulphur dioxide emissions measured

simultaneously with the corresponding particulate tests were plotted and are shown along with a best-fit line in Figure 5. A plot of associated frequency distribution of errors in tonnes per hour is shown in Figure 6.

TABLE 5: MEASURED AVERAGE PARTICULATE EMISSIONS FROM THE INCO 381 m STACK IN TONNES/HOUR

	1973	1974	1975	1976	1977	1978	1979	1980
Emissions in Tonnes/h	2.14	1.96	2.32	0.77	1.50	1.44	0.89	1.57
Standard deviation in Tonnes/h	0.359	0.567	-	0.198	0.212	0.170	0.255	0.099

The broken lines in Figure 5 indicate the limits in the standard error of estimate associated with the prediction of particulate emissions from this straight line relationship. Figure 6 illustrates that the points are scattered around the straight line in a random fashion, suggesting normal distribution of errors. These errors are compounded and consist of measurement errors and random process variations which could cause different relationships between the emissions of these two pollutants. Measurement errors alone could account for as much as 70% of the total error of which 10% could be due to sulphur dioxide determination and 60% to particulate measurements, as discussed earlier.

Despite the errors, or large scatter of individual measurements in Figure 5, this straight-line relationship can be used to either estimate particulate emissions from the knowledge of sulphur dioxide emissions or to assess the process operation when emissions of both pollutants are known. In case the two emissions fall outside the area enclosed by the broken lines in Figure 5, a significant change in the process operation could be suspected.

Some of the emission data used in calculating the straight-line relationship in Figure 5 were obtained shortly after improvements to the electrostatic precipitators had been made, hence, the effects of these improvements on the particulate emissions are included in the relationship (31). These effects are also responsible for the largeness of scatter in Figure 5. In an attempt to further explain this scatter, residuals (difference between the measured and predicted value) from Figure 5 were plotted against years. This plot suggested that a significant drop in emissions may have occurred in 1976; and since the emissions were already normalized for production, this drop could not have resulted from lower production activities in the smelter. Since 1976 the normalized emissions have been steadily increasing until 1980, when they may have been only about 10-20% lower then in 1973.

The straight line relationship in Figure 5 was used to calculate the annual emissions of particulates in the period from 1973 to 1981 from the knowledge of the annual sulphur dioxide emissions (34). In Table 4, the calculated annual particulate emissions and the standard error of estimate are given. The calculated emissions are different from the measurement averages shown in Table 5 because of differences between the production rates during tests and the annual production rates. As a result, the calculated average emissions over the eight year period from 1973 to 1980 are smaller and amount to only about 83% of the average measured values in the same eight year period.

6.2 THE INCO 194 M STACK

This stack is the second largest industrial source of particulate emissions in the Basin. From among the three groups of plants contributing to

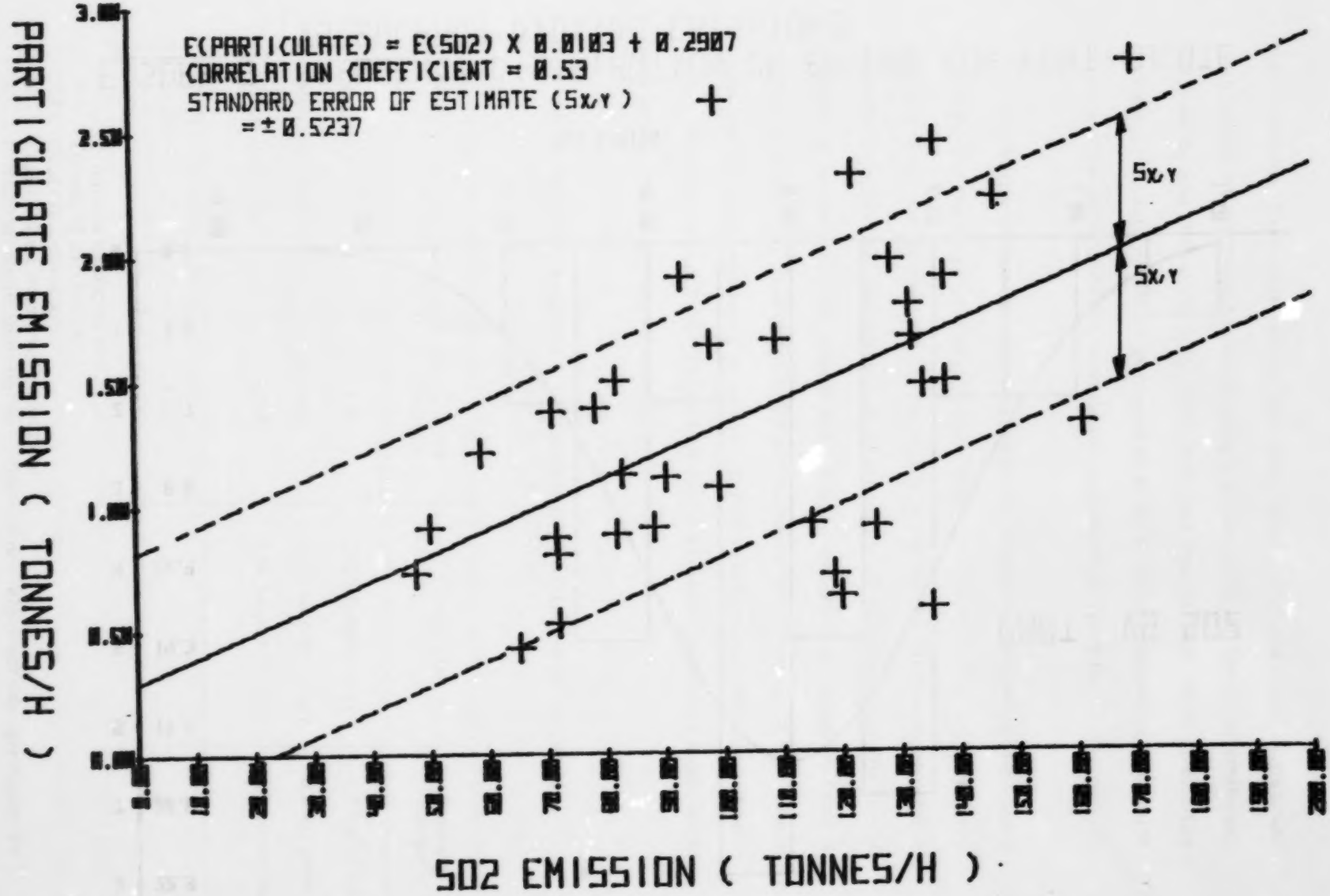

FIGURE 5. TOTAL PARTICULATE VS SULPHUR DIOXIDE EMISSIONS

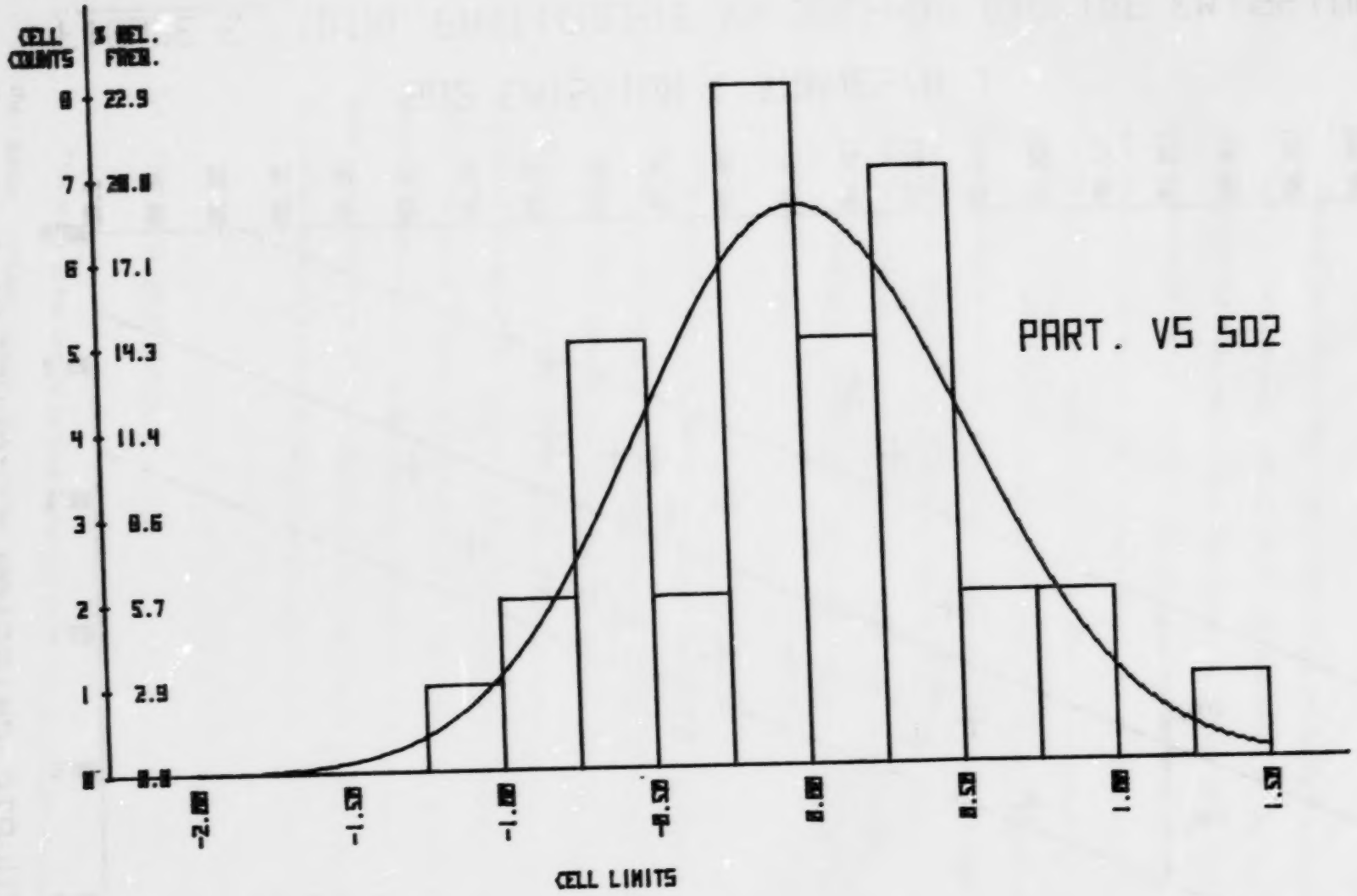

FIGURE 6. FREQUENCY DISTRIBUTION OF ERRORS FOR PARTICULATE VS SULPHUR DIOXIDE EMISSIONS

the emissions from this stack, the acid plants are the least significant contributors of particulates, accounting for about 8% of the total emissions, followed by the nickel refinery with 36% and the I.O.R.P. with 56%. The average particulate emissions as measured in 1976, the corresponding production rates, and the particulate emission factors are shown in Table 6.

Comments on the continuity of emissions from the iron ore recovery plant (I.O.R.P.) and the three acid plants, their intermittency from the nickel refinery plant, and on the production in the plants given in the discussion of sulphur dioxide emissions are also applicable to emissions of particulates. The nickel refinery is an intermittent source and its daily emissions as shown in Table 5 cannot be divided by 24 to obtain the actual hourly emissions since the hourly emissions fluctuate depending on number of heats performed in the day and heat cycle. The emissions in Table 5 represent the total from three heats a day, each heat of four and a half hours duration.

TABLE 6: PARTICULATE EMISSIONS FROM THE INCO 194 M STACK IN TONNES/DAY

Source	Particulate Emissions	Production	Emission Factor^{o}
I.O.R.P.	3.36	2310 tonnes*/day	1.5 kg/tonne
Sulphuric Acid Plants	0.48	2308 tonnes**/day	0.2 kg/tonne
Nickel Refinery	2.68+	3.06 heats***/day	0.9 tonne/heat
Total	6.52+		

* Tonnes of sulphuric acid
** Tonnes of pyrrhotite
*** Heats of converters
o Particulate Emissions/Production
\+ Emissions applicable up to 1980. More electrostatic precipitators installed in 1980.

As far as the accuracy of measurements are concerned, problems were experienced in sampling at both the I.O.R.P. and the nickel refinery plant due to nonideal measurement locations and partial traversing within the horizontal flues. Consequently, the particulate samples collected in these measurements may not be representative of the particulates emitted from these plants. No specific tests were performed at either plant to provide the data required for the estimation of measurement errors, however, these errors could be expected to be larger than the 60% estimated for the 381 m stack.

Particulate emissions from the nickel refinery plant were decreased in 1980 after the installation of additional precipitators in the two converter flues (30). However, no satisfactory emission testing data are presently available to either assess the degree of improvement or to quantify the emissions of particulate pollutants from the new process.

The acid plant emissions in Table 6 include the portion of sulphuric acid which was condensed at the sampling temperature, in accordance with the discussion of particulate emissions from the 381 m stack. No such corrections were applied to the emissions from the other two plants because the data on the condensed portion of the acid was not available..

6.3 THE FALCONBRIDGE 93 M STACK

Particulate emissions from the 93 m stack were measured in 1973 and 1979. The results from 1973 are the only available emission data applicable to the old process at the Falconbridge plant and they suggest fairly high emissions at 19.2 tonnes per day. Unfortunately, since these measurements were done at only one point of the stack cross-section and were short in duration, their accuracy cannot be considered satisfactory.

A more detailed measurement program was executed in 1979, when the new process was on line. The measured emissions were much lower at 2.37 tonnes per day. The error associated with these measurements was estimated at about ± 38%. These emissions were recalculated to include the condensed portion of sulphuric acid.

During the emission measurements, the average feed rate to the roasters was equivalent to 1,703 tonnes per day. The particulate emission factor based on this roaster feed is 1.4 kg of particulates for each tonne of the roaster feed.

6.4 INCO'S LOW LEVEL EMISSIONS

The relative contribution of this source to the total particulate emissions from the Basin is surprisingly larger than in the case of sulphur dioxide emissions. The low level particulate emissions at 1.6 tonnes per day account for about 3.9% of the total particulate emissions in the Basin, while a similar percentage for sulphur dioxide is about 1.1%. It is not known whether this is a result of measurement errors or a reflection of the actual relationship between these two pollutants in low level emissions. As was already indicated in the description of processes and measurement methods, the measurement of low level emissions was a difficult and complex task, which required development of special methods. These methods were at times crude and unproven. For example, in one case, visual observation was used to assign a relative contribution factor to the emissions sampled in a ventilator for each of the two adjacent process units. This factor was then used to calculate emissions from similar units. In other instances, particulate emissions were sampled in large ventilators over long time periods, using methods which had to be simplified in order to make them practical. While these simplications may have a less

significant effect on the accuracy of measurements of gaseous species, e.g. sulphur dioxide, they could cause larger errors in the case of particulates. Hence, it should not be surprising if the measurement error in some tests was even greater than ± 60% as reported by the Inco's smelter process technology group (14).

From among individual smelting processes, the largest contributor to the low level particulate emissions from the smelter are the copper converters, accounting for 64-70% of the total, followed by the multiple hearth roasters with 9-11%, the reverberatory furnaces with 9-11%, the nickel converters with 9-11% and the copper flash furnace with 3% (21,27). Emissions from each of these processes were prorated to include production in all units in the smelter and so prorated emissions are shown in Table 4 (38).

6.5 SIZE OF EMITTED PARTICULATES

Emissions from all processes in consideration are cleaned from a bulk of pollutants, particularly those in particulate form, prior to entering the stacks. Electrostatic precipitators are used in the majority of the plants, except at the sulphuric acid plants where water scrubbers are used. These precipitators are particularly effective in removing particulates of large size, thereby leaving in the emissions mostly particulates with aerodynamic diameters of less than 10 to 15 μm. These small particulates could be airborne and respirable, and, therefore, it is important to characterize them in terms of size and chemical composition.

Commonly used parameters in particle size characterization are the aerodynamic diameters (DP) of the given weight fractions of the particulate

sample. Thus DP 50% of 5 μm means that, in terms of weight, 50% of all emitted particles have aerodynamic diameters equal to or less than 5 μm. In addition to DP 50% or mass median diameter (MMD), cut-off diameters of 15.9% and 84.1% weight fractions of the sample are usually reported, to enable a more complete characterization of the size distribution. Impactor methods, which are frequently used in the size measurements, do not always distinguish between particles larger than about 10 to 15 μm. In order to obtain the size information in this range, the measured data must be extrapolated, which in turn may accentuate small measurement errors to quite large differences in the extrapolated results. This source of error must be recognized when interpreting the sizing data.

Particulate sizes were measured at the Inco 381 m stack on several occasions and at the Falconbridge's 93 m stack once. No measurements were performed at the other sources. Both in-stack and in-plume measurements were carried out at both of these stacks.

6.5.1 PARTICULATE SIZE MEASUREMENTS AT THE INCO 381 M STACK

The in-stack particulate size measurements at the 381 m stack were done over the period of eight years, and the results of these measurements are given in Table 7.

The available test data from 1973 do not contain the weights of large particles retained in the inlet portions of the impactor samplers, resulting in too small mass median diameters, as shown in Table 7. However, if the data from 1976 are truncated to exclude the inlet portions of the samples, the mass median

TABLE 7: SUMMARY OF PARTICLE SIZE MEASUREMENTS AT THE INCO 381 M STACK

Year	No of Tests	DP 50% μm	Dp 15.9% μm	DP 84.1% μm	% of Sample included
1973*	2	1.9 - 2.9*			50%
1976	3	6.8 - 7.4	0.9 - 1.3	16 - 47	100%
1977	1	8	1.2	48	100%
1978	3	3.5 - 5.5	0.65 - 0.89	19 - 36	100%
1980	3	3.3 - 6.4	0.53 - 0.72	23 - 60	100%

* Incomplete sample yielding a nonrepresentative result.

diameters of the truncated samples are comparable to those from the 1973 program. This suggests that the size of particulates in 1973 and 1976 may have been similar. The particulate size distributions in 1976 and 1977 were also similar. Tests in 1978 and 1980 resulted in slightly smaller particulates. Measurements in 1978 were particularly exhaustive, and the results from that year can be considered accurate and representative of emissions from this stack (18).

6.5.2 PARTICULATE SIZE MEASUREMENT AT THE FALCONBRIDGE 93 M STACK

The size measurement tests at this source were performed in 1979, and the particulates were found to be somewhat smaller than in the Inco 381 m stack. Since similar measurement methods were used at both sources, the differences in the results must be attributed to processes and process equipment, including the electrostatic precipitators at the two smelters. The results obtained at the Falconbridge stack are given in Table 8.

TABLE 8: SIZE OF PARTICULATES EMITTED FROM THE FALCONBRIDGE 93 M STACK

No of Tests	DP 50% μm	DP 15.9% μm	DP 84.1% μm	% of Sample Included
3	2.3 - 4	0.44 - 0.66	10 - 33	100%

In-plume measurements were also carried out at Falconbridge, and no significant differences between the in-plume and in-stack size measurement results for most metals of interest were recorded (19, 23).

7. EMISSIONS OF METALS

Emissions of metals in the Sudbury Basin follow the trend established for particulates, thus, on the average the largest source of nickel, lead and arsenic emissions is the Inco 381 m stack, followed by the Inco 194 m stack, the Falconbridge 93 m stack, and Inco's Copper Cliff smelter building. However, in terms of particulate composition, the 194 m stack is by far the most significant source of copper and nickel. Even in terms of absolute quantities, this source used to emit almost as much of these two metals as the 381 m stack prior to installation of additional precipitation capacity at the nickel refinery in 1980. (No satisfactory emission data applicable to the present operation at the refinery are available). Another significant source of copper is Inco's Copper Cliff smelter building, contributing about 37% of all copper emissions in the Basin. Iron was mainly emitted from Inco's two 45 m stacks until the pellet plant shutdown in 1980.

Not all metals were determined in all emission measurements for the Sudbury sources, however, the major species which, on the average, exceeded both one percent content of total particulate and the emission rate of 10 kg/h from the 381 m stack over the eight year period from 1973 to 1980, were quantified at all sources, and their emissions are listed in Table 9.

The iron, copper, and nickel emissions from the 381 m stack were calculated from correlations with sulphur dioxide, while lead and arsenic emissions were averaged from three or more valid emission measurement tests. Emissions of metals other than those in Table 9, including cobalt, selenium, mercury, chromium, manganese etc., accounted for less than 1% of particulate emissions, as discussed under individual sources.

TABLE 9: AVERAGE EMISSIONS OF METALS FROM MAJOR SUDBURY SOURCES IN KILOGRAMS/HOUR

Source	Iron	Copper	Nickel	Lead	Arsenic	Total	Percent of Contribution
Inco 381 m Stack	113 Sx,y=109.5*	28 Sx,y=19.8*	26 Sx,y=16.4*	21 σ=8.6**	13 σ=5.5**	201	53.5
Inco 194 m Stack°	73	19.5	26.2	0.7	0.4	119.8	31.9
Inco smelter building low level	8	28	3.5	0.1	0.01	39.6	10.5
Falconbridge 93 m Stack	11	1	1	1.5	0.8	15.3	4.1
Inco two 45 m Stacks	(268.8)+						
Total°	205	76.5	56.7	23.3	14.2	375.7	

* Sx,y = standard error of estimate
** σ = standard deviation
° Emissions measured in 1977, inapplicable after the installation of new electrostatic precipitators at the nickel refinery in 1980.
\+ Emissions inapplicable after the pellet plant shutdown in 1980. Excluded from the total.

Metals were analysed in almost every sample collected in the course of particulate emission measurements at all Sudbury sources. Analyses were frequently performed by at least two laboratories, Falconbridge's or Inco's and the Ministry of the Environment's. In the first sampling campaign at the 381 m stack, a third independent laboratory was engaged to analyse selected samples for cross-checking purposes. This was justified on the basis of uncertainties and novelties involved in early analytical work; however, when the proper procedures were well established and the approximate levels of metals in the emissions became known, the number of laboratories involved in any given measurement program was decreased. The number of metals analysed in particulate samples was also decreased to include only major constituents.

Some metals of interest, mercury, arsenic, lead and selenium, are volatile and their collection in the sampling stage was not always assured. Thus, if the particulate sample was analysed for any of these four species with no modification in the sampling procedure to ensure their quantitative collection, the obtained results were in error and inconsistent with expectations. In the case of the 381 m stack, these uncertainties were removed by performing specially designed emission tests in 1980, and this program resulted in the most reliable emission data for these metals from this source.

The metal emissions from the 381 m stack depend on production and increase proportionately with the sulphur dioxide emissions. This dependence and a relative abundance of pertinent data enabled development of correlations, which were then used to estimate annual emissions of copper, nickel, and iron from the 381 m stack. An insufficient number of tests at other Sudbury sources was the reason that no similar correlations for those sources were produced.

7.1 THE INCO 381 M STACK

The best estimates of average hourly emissions of major metals from the 381 m smokestack in the period from 1973 to 1980 are shown in Table 10.

The iron, copper, and nickel emissions shown in this Table were calculated on the basis of yearly sulphur dioxide emissions from straight line correlations between the measured emissions of each metal and sulphur dioxide (the measured emissions of these metals are shown in Table 11). The straight lines with the limits of standard errors of estimates and the associated frequency distribution of errors graphs are shown in Figures 7 through 12.

TABLE 10: AVERAGE EMISSIONS OF FIVE MAJOR METALS FROM THE INCO 381 M STACK IN KILOGRAMS/HOUR

Year	Iron*	Copper*	Nickel*	Lead	Arsenic
1973	127	31	29	21	11
1974	163	39	37	29	10
1975	166	39	38	23	23
1976	169	39	39	12	15
1977	156	37	35	18	8
1978	52	14	12	10	-
1979	23	8	6	-	-
1980	89	22	21	34	10
1981	73	19	17		
Average	113	28	26	21	13

*Emissions calculated from regression lines

The straight-line correlations were selected after several linear and nonlinear functions were examined for the goodness of fit through the measured data points plotted on graphs. Among the linear functions, polynomials of up to fifth order were studied. In comparison to the straight lines, the polynomials of higher order were either very similar to the straight lines or unsuitable in that they overfit the data points. Among the nonlinear functions, the exponential correlations resulted in improved correlation coefficients, but also in poorer distribution of errors than the straight-line correlations. The average emissions of the three metals calculated from the exponential correlations over the eight years from 1973 to 1980 were lower than the corresponding emissions calculated from either the straight lines or the average measured values. However, when the errors of estimates were taken into account, no significant difference in the values estimated from either function could be found. In this situation a choice of the mathematically simpler straight lines was justified. The estimates of emissions from the straight-line correlations as shown in Table 10 are more representative of the actual emissions than the mathematical averages of measured emissions shown in Table 11 because they were normalized with respect to the production rates.

The scatter of individual data points about the lines in Figures 7, 9 and 11 is admittedly large, but also understandable in the light of variations in particulate composition, process operation, suiphur dioxide emissions, and the measurement errors which could all contribute to its magnitude. Errors are associated with the estimates from these lines and they must be taken into account when interpreting the calculated emissions. Distribution of errors of estimates about these straight lines seem skewed, suggesting that a lognormal analysis could be appropriate. It must be also ensured that no significant changes in the process have taken place, if the emissions beyond 1980 are calculated from these straight lines. Better correlations with less scatter were obtained in the case of metals versus particulates, indicating consistency in the chemical analyses of the emission samples.

Attempts to correlate arsenic and lead emissions with sulphur dioxide were unsuccessful in that their emissions were inversely proportional. One of the possible reasons for this unexpected result is nonquantitative collection of these species during particulate emission measurements. A special sampling method for arsenic, selenium and mercury was employed in the 1980 sampling program, but the results did not indicate large differences from the past programs.

Measured average emissions of all species in particulate form, including metals, are shown in Table 11.

In-plume measurements of metal emissions from the 381 m stack were performed simultaneously with the in-stack measurements in 1977 (11, 15). The results from these tests were comparable, except for arsenic with higher emissions measured in a colder plume. A similar comparison attempted in 1980 resulted in larger discrepancies, with the in-plume method yielding several times

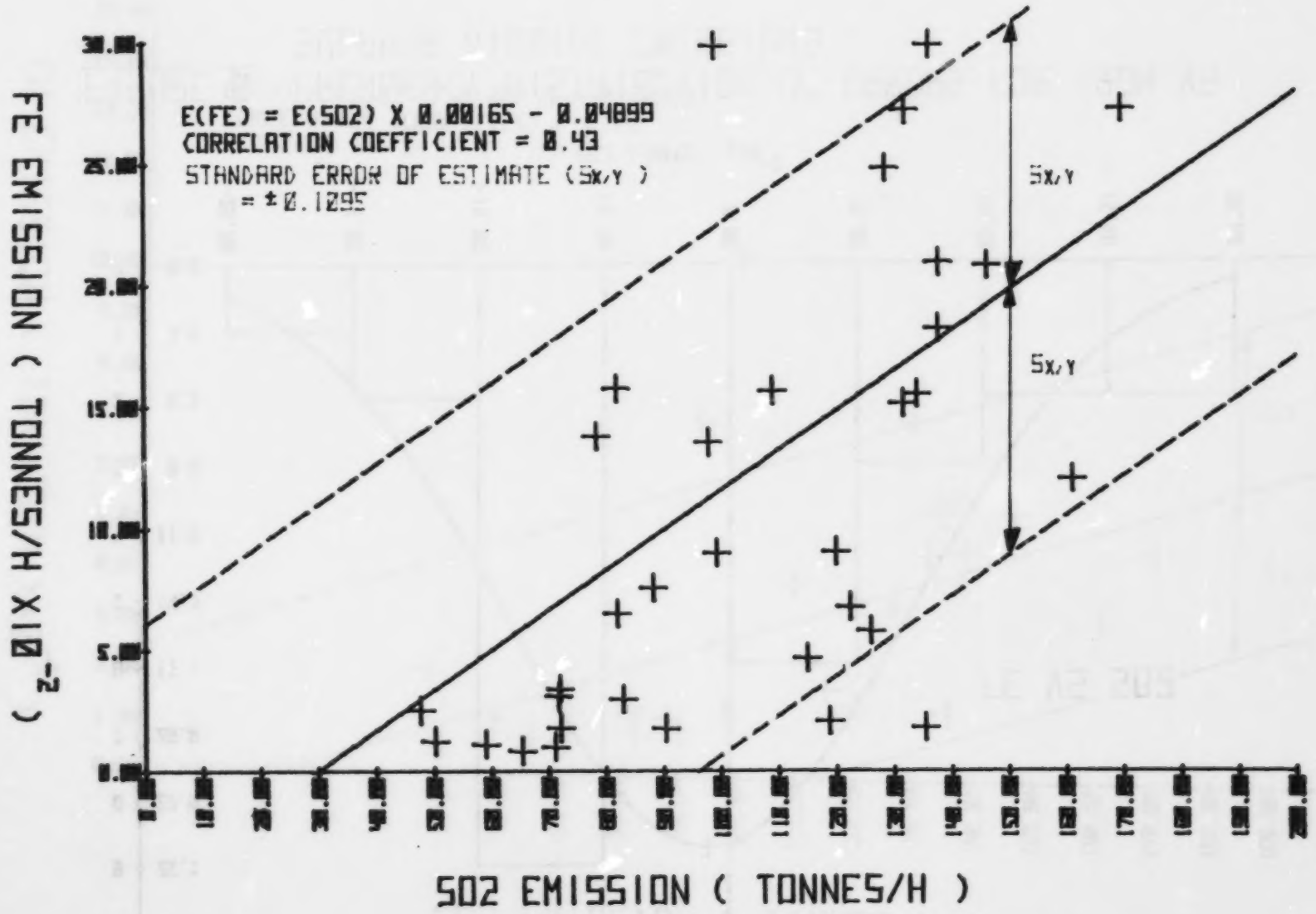
E(FE) = E(SO2) X 0.00165 - 0.04899
CORRELATION COEFFICIENT = 0.43
STANDARD ERROR OF ESTIMATE (Sx/Y)
= ± 0.1295
Sx/Y
Sx/Y
FE EMISSION (TONNES/H X 10^-2)
SO2 EMISSION (TONNES/H)

FIGURE 7. IRON VS SULPHUR DIOXIDE EMISSIONS

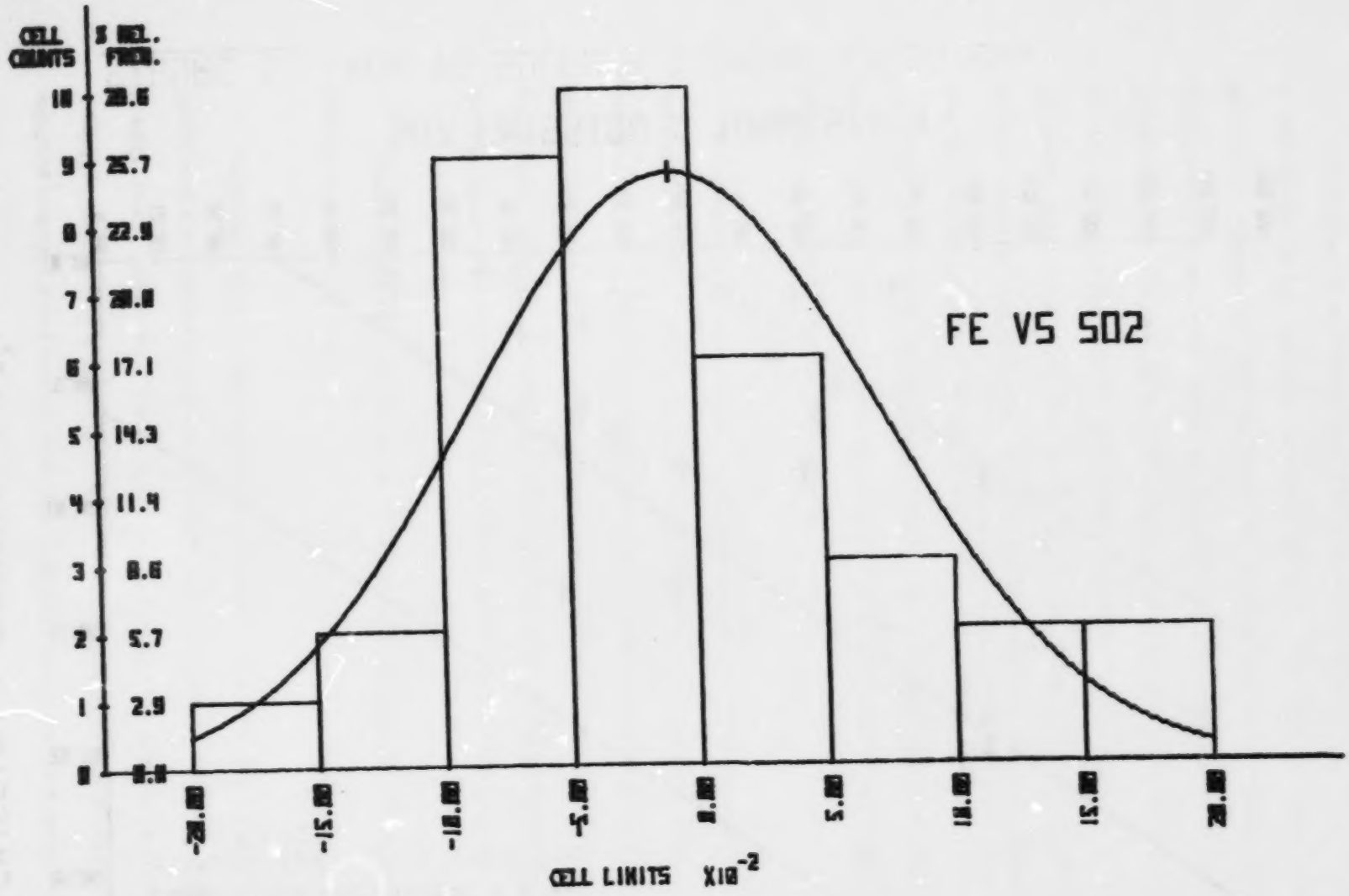

FIGURE 8. FREQUENCY DISTRIBUTION OF ERRORS FOR IRON VS SULPHUR DIOXIDE EMISSIONS

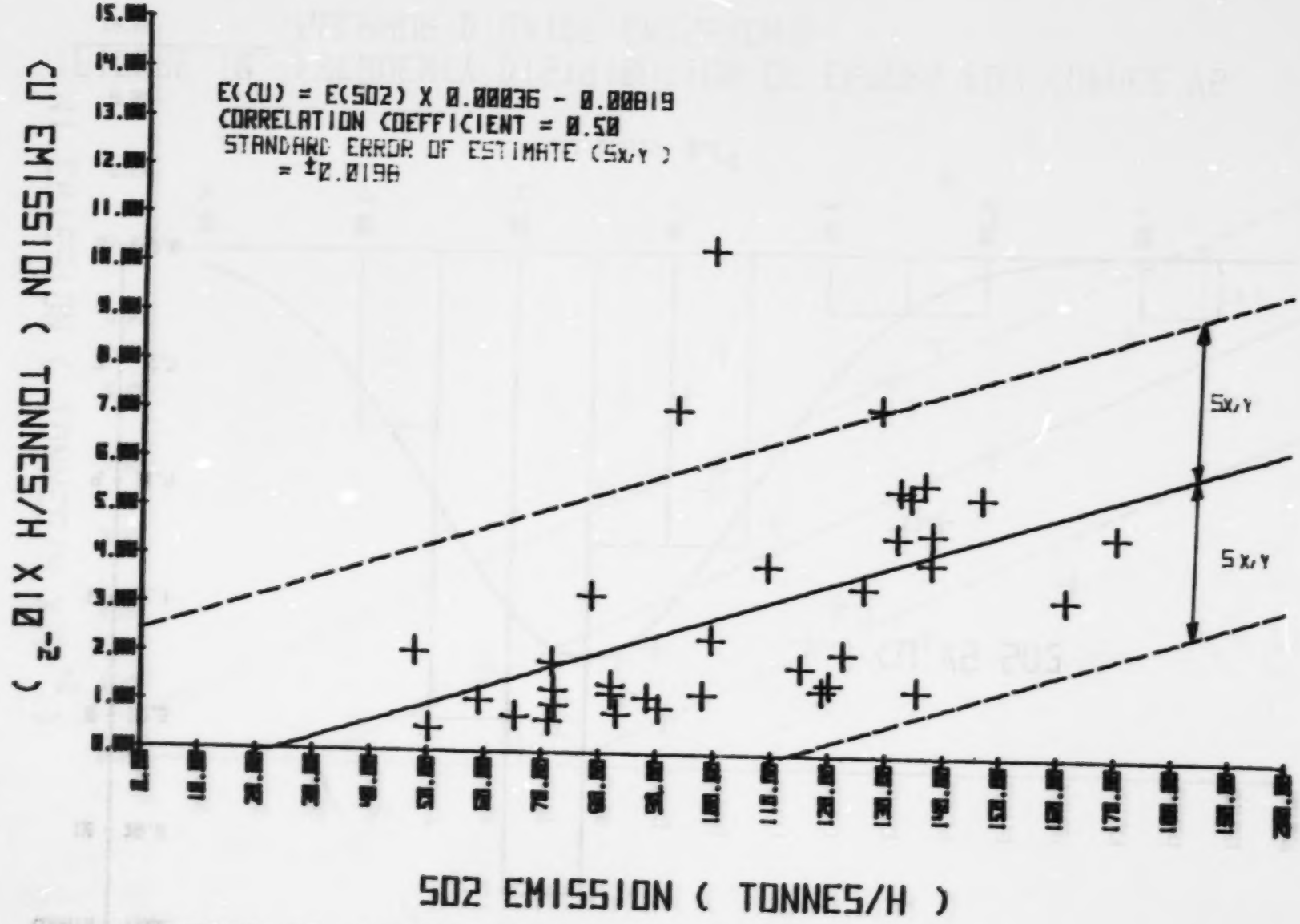

FIGURE 9. COPPER VS SULPHUR DIOXIDE EMISSIONS

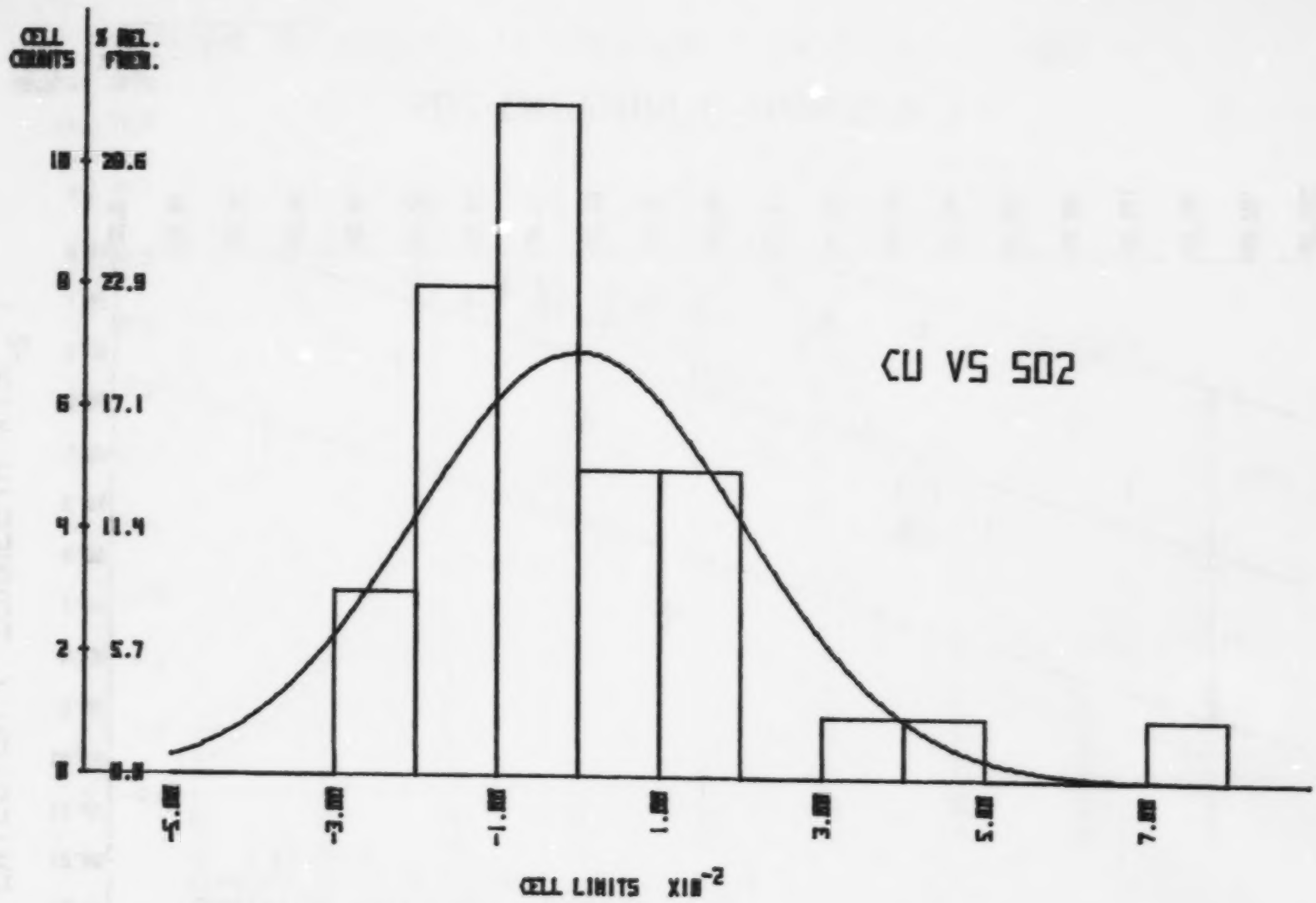

FIGURE 10. FREQUENCY DISTRIBUTION OF ERRORS FOR COPPER VS SULPHUR DIOXIDE EMISSIONS

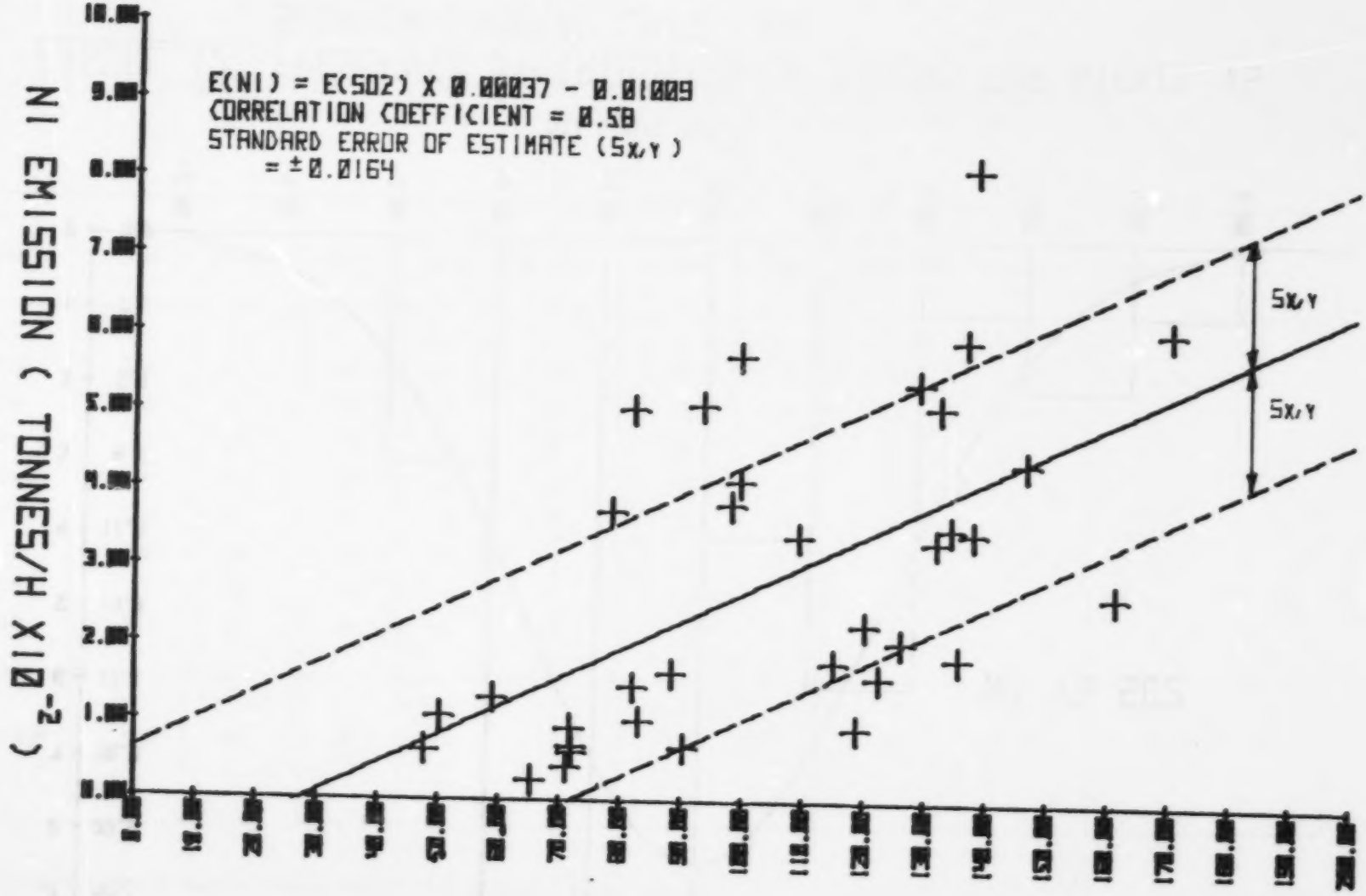
E(NI) = E(SO2) X 0.00037 - 0.01009
CORRELATION COEFFICIENT = 0.58
STANDARD ERROR OF ESTIMATE (Sx,y)
= ±0.0164
Sx,y
Sx,y
NI EMISSION (TONNES/H X 10-2)
SO2 EMISSION (TONNES/H)

FIGURE 11. NICKEL VS SULPHUR DIOXIDE EMISSIONS

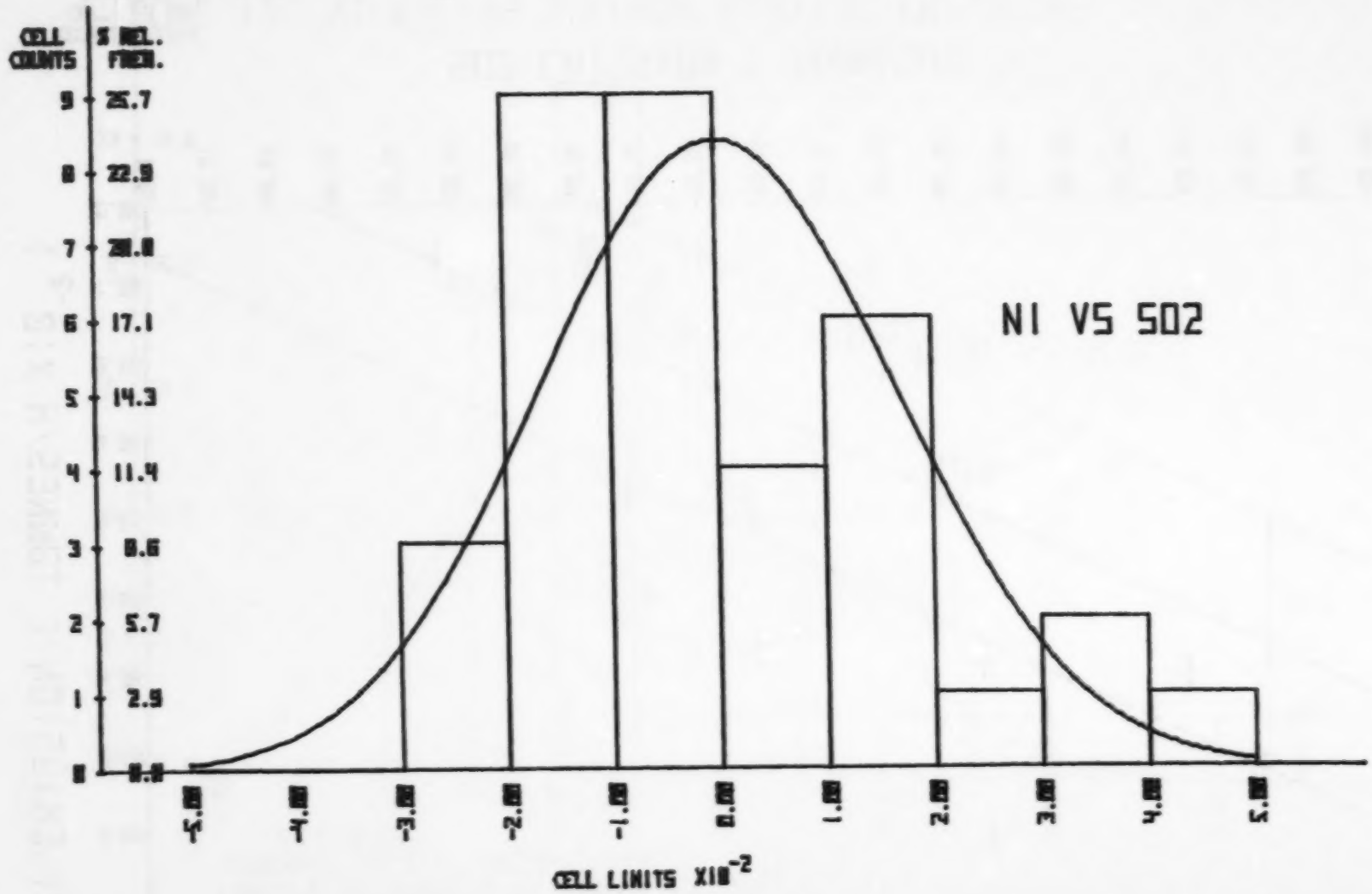

FIGURE 12. FREQUENCY DISTRIBUTION OF ERRORS FOR NICKEL VS SULPHUR DIOXIDE EMISSIONS

TABLE 11: AVERAGE MEASURED EMISSIONS OF METALS AND OTHER PARTICULATE POLLUTANTS FROM THE INCO 381 M STACK IN KILOGRAMS/HOUR

Year	Iron	Copper	Nickel	Lead	Aluminum	Arsenic	Zinc	Chromium	Selenium	Cadmium
1973	228	65	48	21	18	11	6	6	6	4
1974	258	55	55	29	6	10	7	15	6	5
1975	64	27	15	23	4	23	11	0.5	6	0.8
1976	74	25	22	12	3	15	7	0.2	-	1
1977	142	41	33	18	9	8	11	2	-	1
1978	80	20	20	10	-	-	1	-	-	-
1979	31	13	12	-	-	-	-	-	-	-
1980	147	47	44	34	19	10	6	2	3	2
Average	128	37	31	21	10	13	7	4	5	2

* Emission rates of mercury were measured in 1980 at 7 grams per hour.

TABLE 11 (CONTINUED): AVERAGE MEASURED EMISSIONS OF METALS AND OTHER PARTICULATE POLLUTANTS FROM THE INCO 381 M STACK IN KILOGRAMS/HOUR

Year	Bismuth	Beryllium	Manganese	Tin	Antimony	Strontium	Silver	Vanadium	Lithium	Silicon
1973	3	2	1	0.8	0.6	0.4	0.3	0.2	0.1	109
1974	3	-	2	-	-	-	-	-	-	19
1975	2	-	0.2	-	-	-	-	-	-	14
1976	1	-	0.1	-	-	0.1*	0.1	0.1	0.6	-
1977	5	-	0.4	-	-	-	-	1.0	-	32
1978	-	-	-	-	-	-	-	-	-	-
1979	-	-	-	-	-	-	-	-	-	-
1980	-	-	0.3	-	-	-	-	-	-	-
Average	3	2	0.7	0.8	0.6	0.5	0.2	0.4	0.3	43

TABLE 11 (CONTINUED): AVERAGE MEASURED EMISSIONS OF METALS AND OTHER PARTICULATE POLLUTANTS FROM THE INCO 381 M STACK IN KILOGRAMS/HOUR

Year	Magnesium	Boron	Calcium	Tellurium	Cobalt	Molybdenum	Titanium	Potassium	Sodium
1973	15	10	9	0.3	-	-	-	-	-
1974	5	-	5	-	2.0	4	-	-	-
1975	1	-	3	-	0.3	-	-	-	-
1976	-	-	-	-	0.1	0.1	1	4	20
1977	6	-	7	-	1.0	-	-	3	2
1978	-	-	-	-	-	-	-	-	-
1979	-	-	-	-	0.3	-	-	-	-
1980	4	-	-	-	2.0	-	-	-	-
Average	6	10	6	0.3	0.9	2	1	3	11

lower emissions for most metals. Arsenic was again an exception, with much higher emissions measured by the in-plume method.

From among the five major metals, iron, copper, and nickel tend to be emitted in larger sizes, with mass medium diameters of more than 13 um, while the more volatile lead and arsenic are much smaller in size, with MMDs of about 1 um (35).

7.2 THE INCO 194 M STACK

The iron ore recovery plant and the nickel refinery are the only plants contributing metals to the 194 m stack emissions (the acid plant emissions are essentially metal-free). The combined emissions of iron, nickel and copper from these two processes are as large as those from the 381 m stack. Iron is largely emitted from the iron ore recovery plant, while copper and nickel are mostly contained in the nickel refinery emissions. In-stack emission measurements at all plants - iron ore recovery, nickel refinery and sulphuric acid plants - were performed only on one occasion in 1976, consequently, no dependence of emissions on production can be illustrated. Rather, the production rates during the measurements, as shown in the relevant Inco reports (9, 10), will be given. In addition to these in-stack measurements from 1976, there are 1979 in-plume emission data available for comparison of the two methods; this is illustrated in Table 12 (28).

As will be discussed later, the in-stack measurements were not performed according to strict testing procedures, and the results could be in error. In the case of copper and nickel they are probably too low. The in-stack and the in-plume measurements were not conducted simultaneously, but since the two processes were operated at similar production rates during these measurements,

TABLE 12: COMPARISON OF IN-STACK AND IN-PLUME AVERAGE EMISSIONS IN TONNES/DAY

Method	Sulphur Dioxide	Iron	Copper	Nickel	Lead	Arsenic
In-stack 1976	150	1.8	0.5	0.6	0.01	0.01
In-plume 1979	190	0.8	0.2	0.5	0.01	0.04
$\frac{\text{In-stack}}{\text{In-plume}}$	0.8	2.2	2.5	1.2	1	0.25

the emissions obtained by the two methods can be compared. As already discussed, the in-plume method produced relatively lower emissions of iron, copper and nickel, and higher emissions of arsenic.

Emissions of metals (in-stack measurements) from the iron ore recovery and nickel refinery plants are shown in Table 13. Emissions from the iron ore recovery plant are continuous, and the corresponding values from Table 13 can be prorated to any time period. As far as representativeness of emissions is concerned, similar comments to those made in the discussion of particulate emissions from this plant apply. The sampling method employed in the emission measurements was not rigorous enough to ensure collection of representative samples and large errors in the emissions as shown in Table 13 are possible.

The emissions from the nickel refinery may also be in error and are probably too low. These emissions are not continuous; they originate in nickel converting, which is a batch process. In calculating the emissions in Table 13, it was assumed that 3.06 batches of 70 tonnes of charge, each of 4.5 hours duration, were processed in a day. In order to obtain approximate actual

TABLE 13: EMISSIONS OF METALS FROM THE IRON ORE RECOVERY AND NICKEL REFINERY PLANTS IN KILOGRAMS/DAY

Plant	Iron	Copper	Nickel	Lead	Arsenic	Selenium	Cobalt
Iron Ore Recovery	1694	7	48	1	1	0.1	0.9
Nickel Refinery	67	461	581	15	8	3	11
Total	1761	468	620	16	9	3	12

TABLE 13 (CONTINUED): EMISSIONS OF METALS FROM THE IRON ORE RECOVERY AND NICKEL REFINERY PLANTS IN KILOGRAMS/DAY

Plant	Chromium	Manganese	Silver	Cadmium	Aluminum	Magnesium	Calcium
Iron Ore Recovery	8	3	0.7	0.2	-	-	-
Nickel Refinery	7	1	5	2	1	0.5	1
Total	15	4	6	2	-	-	-

emissions per batch, the emissions in Table 13 must be divided by 3.06. Metal emissions are not uniform throughout the batch and they change in each of the three cycles - charging, blowing and reduction. In comparison to nonvolatile metals such as iron, copper, and nickel, which are mostly emitted during charging and blowing, half of the more volatile lead and arsenic are emitted during reduction.

7.3 LOW LEVEL EMISSIONS FROM THE INCO SMELTER BUILDING

Except for copper, which is emitted in considerable quantities in unconfined or fugitive emissions from the copper converters, the low level emissions from the Inco smelter building are relatively small contributors to the total emissions of metals in the Sudbury Basin. The low level metal emissions as measured simultaneously with the particulates were prorated to a typical production of nickel bessemer matte and blister copper (12, 14, 17, 20, 21) and the prorated emissions from various process steps are shown in Table 14.

The accuracy of these emission measurements was low, for reasons already discussed under particulate emission measurements, hence, the degree of representativeness of emissions in Table 14 is not known. However, they are the best measurements presently available and at least accurate enough to confirm what was suspected before; that is, in comparison to the other sources discussed in this report, the low level emissions from the smelter building are not a significant contributor to the total metal emissions in the Sudbury Basin, with the exception of copper.

7.4 THE FALCONBRIDGE 93 M STACK

Emissions of metals from the Falconbridge smelter were measured with

TABLE 14: AVERAGE LOW LEVEL EMISSIONS OF METALS IN KILOGRAMS FROM THE INCO SMELTER AT COPPER CLIFF DURING TYPICAL PRODUCTION RATES OF NICKEL BESSEMER MATTE AND BLISTER COPPER IN KILOGRAMS/DAY

Process	Iron	Copper	Nickel	Lead	Arsenic	Cobalt	Zinc	Cadmium	Selenium
Copper converters	82	602	50	-	-	1.0	-	-	-
Nickel converters	27.9	19	22.4	0.2	0.1	0.86	0.2	0.01	0.03
Nickel reverberatory furnaces	27.2	5.2	8.0	T* -	T* -	0.31	T* -	-	T* -
Multiple hearth roasters	57.0	4.7	12.1	T**	T**	0.41	T**	T**	T**
Copper flash furnace	5.7	8.2	0.3	1.5	0.01	0.01	0.61	0.3	-
Total	199.8	639.1	92.1	1.7	0.11	2.6	0.81	0.31	0.1

T* Total Emissions of Lead, Zinc, Cadmium, Bismuth, Arsenic, and Selenium estimated at 1 Kg/D (17)

T** Total of Lead, Zinc, Cadmium, Bismuth, Arsenic, and Selenium emissions - 1.33 Kg/D (21)

both old and new smelting processes on line. The results suggested that there was a significant decrease in emissions of at least the major metals occurred after the installation of the new process in 1978. Admittedly, the measurement methods prior to 1978 were not as accurate as the multipoint in-stack sampling executed at the 93 m stack in 1978, and the results may be in error. In 1973, an in-stack-single-point grab sample was taken (23), while a comprehensive in-plume method was employed in 1977 (15). The in-plume method can also underestimate the emissions of nonvolatile metals, as discussed under the metal emissions from the 194 m stack; hence, the emissions of these metals from the old process could have been even higher than measured and shown in Table 15. In spite of this possible underestimation, it is clear from Table 15 that the metal emissions from the smelter have been significantly reduced after the installation of the new process.

There is a degree of similarity in the size distribution of metals emitted from the Falconbridge 93 m and Inco 381 m stacks; iron, nickel and copper are emitted at a larger size than the more volatile lead and zinc. However, in terms of absolute size, all metal particles from the Falconbridge stack were smaller, with the mass median diameters for iron, nickel and copper ranging from 3.0 to 7.0 micrometers (26) and 0.5 to 0.9 micrometers for lead and zinc; comparable values at the Inco 381 m stack were larger than 13.0 micrometers for non-volatile metals, and 0.5 to 4.0 micrometers for lead and zinc.

No correlation or dependence of metal emissions on production at the Falconbridge smelter can be shown because of an insufficient number of measurements.

TABLE 15: EMISSIONS OF METALS FROM THE FALCONBRIDGE 93 M STACK IN KILOGRAMS/DAY

Method	Iron	Copper	Nickel	Lead	Arsenic	Selenium	Zinc	Aluminum	Manganese	Cadmium	Magnesium	Calcium
In-stack 1979	270	31	25	37	18	-	8	20	0.3	6	11	9
Standard Deviation	202	25	19	21	16	-	3	23	0.2	3	8	6
In-plume 1979	115	28	19	30	131	-	5	29	0.7	2	-	-
In-plume 1977	2310	384	269	140	441	8	56	-	13	-	-	-
In-stack 1973	809	920	1240	140	-	-	-	-	-	-	-	-

8. EMISSIONS OF SULPHURIC ACID

The difficulties inherent in determining sulphuric acid in emissions from mineral processing were already discussed in the description of sampling methods. Consequently, a possibility exists that some of the measured emissions, with the exception of acid plants, are in error. This possibility was recognized after detailed analyses of test data, which were not always consistent with either theoretical expectations or other measurement programs (4, 11). In most instances, inconsistencies were traced to improper sampling conditions and low sampling temperatures which allowed condensation of sulphuric acid on particulates and, hence, possible formation of artifact sulphates. Sulphates including those of artifact origin, were excluded from the calculations of acid emissions, and so calculated emissions could have been underestimated. The total sulphate emissions from the 381 m stack were in some tests as large as the sulphuric acid emissions (16). There are no similar interferences in the acid plant emissions, and these emissions can be considered accurate.

Depending on the source, the acid emissions account for 0.4 to 3.0 percent of the total sulphur dioxide and acid emissions. In the Inco 381 m stack and the Falconbridge 93m stack emissions, these percentages range from 0.4 to 1.4, while in the Inco 194 m stack, the percentage is higher at about 3%. The largest source of acid emissions is the 381 m stack, contributing about 77% to the total of 25.9 tonnes of acid per day from all major sources in Sudbury, as indicated in Table 1.

Hourly emissions from all sources are shown in Table 16.

TABLE 16: ACID EMISSIONS FROM MAJOR INDUSTRIAL SUDBURY SOURCES IN TONNES/HOUR

Year	Inco 381 m Stack	Inco 194 m Stack	Inco Smelter Low Level	Falconbridge 93 m Stack
1973	0.63			
1974	1.04			
1975	1.66			
1976	-			
1977	0.37	0.19		
1978	-		0.01	
1979	0.74			0.05
1980	0.52			
Average	0.83	0.19	0.01	0.05

Acid emissions from the 381 m smokestack were not always consistent with sulphur dioxide emissions. Sulphur dioxide emissions were higher in 1977 than in 1973 when acid emissions were higher, thus causing problems in developing a satisfactory correlation between the emissions of these two species. However, data from 1973 through 1976 suggest that the emissions of these two species increase simultaneously (1 through 7).

From among the three processes contributing to the 194 m stack emissions, the acid plants are the most significant source of acid. Combined emissions from the three acid plants were 0.1325 tonnes per hour, representing about 71% of the total to the stack, followed by the nickel refinery with 0.044 tonnes per hour, and the iron ore recovery plant with 0.01 tonnes per hour. Emissions from the nickel refinery are not continuous, and the daily maximums occur in three one-hour periods.

Determination of sulphuric acid in the Inco smelter low level emissions was not complete in that no satisfactory measurements were performed at the nickel and the copper converters. No sulphuric acid was found in the fugitive emissions from the roasters. The emissions listed in Table 16 include only the reverberatory furnaces and the copper flash furnace.

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